DECLARATION

In the matter of U.S. Patent Application Ser. No. 09/830,923 in the name of Keiichi IMAMURA, et al.

I, Kyoko IMAMURA, of Kyowa Patent and Law Office, 2-3, Marunouchi 3-Chome, Chiyoda-Ku, Tokyo-To, Japan, declare and say:

that I am thoroughly conversant with both the Japanese and English languages; and,

that the attached document represents a true English translation of Japanese Patent Application No. 1998-313688 filed on November 4, 1998.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Dated: May 27, 2005,

Kyoko Imamura

Kyoko IMAMURA

10-313688

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Application Date: November 4, 1998

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International Patent Classification: A01N 43/40

Title of the Invention: PICOLINAMIDE DERIVATIVE AND HARMFUL ORGANISM CONTROL AGENT FOR PLANT COMPRISING SAID PICOLINAMIDE DERIVATIVE AS ACTIVE COMPONENT

Number of Claim(s): 9

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[Document Name] SPECIFICATION 10-313688

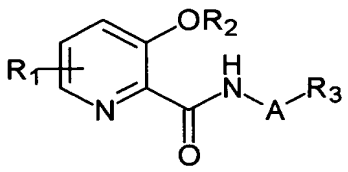
[Title of Invention] PICOLINAMIDE DERIVATIVE AND HARMFUL ORGANISM CONTROL AGENT FOR PLANT COMPRISING SAID PICOLINAMIDE DERIVATIVE AS ACTIVE COMPONENT

[Claims]

[Claim 1]

A picolinamide derivative represented by formula (1) or a salt thereof:

[Chem. 1]



(1)

wherein

[A represents a bond or an optionally substituted alkylene chain;

R_1 represents a hydrogen atom, one, two or more groups, which may be the same or different, selected from the group consisting of, alkoxy, and haloalkoxy;

R_2 represents a hydrogen atom, optionally substituted benzyl, optionally substituted alkyl or optionally substituted alkanoyl; and

R_3 represents a hydrogen atom, optionally substituted cycloalkyl, optionally substituted cycloalkenyl, optionally substituted aryl or an optionally substituted heterocyclic group (provided that the case where R_1 represents a hydrogen atom, A represents a bond or a methylene chain, and R_3 represents phenyl or cyclohexyl, and the case where A represents an alkylene chain and R_3 represents a hydrogen atom are excluded)].

[Claim 2]

The picolinamide derivative represented by formula (1) or a salt thereof according to claim 1, wherein

[A represents a bond or an alkylene chain having 1 to 12 carbon atoms;

R_1 represents a hydrogen atom, one, two or more alkoxy groups having 1 to 4 carbon atoms or haloalkoxy having 1 to 4 carbon atoms;

R_2 represents a hydrogen atom, an optionally substituted benzyl group, optionally substituted alkyl having 1 to 4 carbon atoms or optionally substituted alkanoyl having 1 to 4 carbon atoms;

R_3 represents a hydrogen atom, optionally substituted cycloaklyl having 3 to 12 carbon atoms, or optionally substituted cycloalkenyl having 3 to 12 carbon atoms [wherein the substituent is one, two or more groups, which may be the same or different, selected from the group consisting of a halogen atom, cyano, nitro, carboxyl, hydroxyl, and phenyl (phenyl may be substituted by one, two or more substituents, which may be the same or different, selected from the group consisting of a halogen atom, cyano, nitro, amino, alkylamino, alkanoylamino, alkyl having 1 to 4 carbon atoms, haloalkyl having 1 to 4 carbon atoms and haloalkoxy having 1 to 4 carbon atoms), alkyl having 1 to 4 carbon atoms, haloalkyl having 1 to 4 carbon atoms, or alkoxycarbonyl having 1 to 4 carbon atoms],

optionally substituted monocyclic or polycyclic 3- to 12-membered aryl or 3- to 12-membered heterocyclic group [the substituent is one, two or more groups, which may be the same or different, selected from the group consisting of a halogen atom, cyano, nitro, amino, hydroxyl, formyl, carboxyl, carbamoyl, or thiocarbamoyl;

straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulfinyl, or alkylsulfonyl, having 1 to 6 carbon atoms;

straight-chain or branched alkenyl or alkenyloxy having 2 to 6 carbon atoms;

haloalkyl, haloalkoxy, haloalkylthio, haloalkylsulfinyl, or haloalkylsulfonyl, wherein said groups are straight-chain or branched groups having 1 to 6 carbon atoms that each have 1 to 13 halogen atoms which may be the same or different;

straight-chain or branched haloalkenyl, haloalkenyloxy having 2 to 6 carbon atoms that each have 1 to 13 halogen atoms which may be the same or different,

acylamino, N-acyl-N-alkylamino, alkylamino, dialkylamino, alkylcarbonyl, alkylcarbonyloxy, alkoxycarbonyl, alkylsulfonyloxy, hydroxyiminoalkyl or alkoxyiminoalkyl, wherein said groups each have straight-chain or branched alkyl having 1 to 6 carbon

atoms;

alkylene, dioxyalkylene, polyoxaalkylene, or cycloalkyl having 3 to 6 carbon atoms wherein said groups may be substituted by one, two or more substituents selected from the group consisting of a halogen atom, straight-chain or branched alkyl having 1 to 4 carbon atoms, and straight-chain or branched haloalkyl having 1 to 4 carbon atoms, which has 1 to 9 halogen atoms which may be the same or different, and are present as a chain which is substituted in its both ends at adjacent positions on the ring to form a ring]; and

aryl, aryloxy, arylthio, arylsulfinyl, arylsulfonyl, arylamino, arylalkyl, arylalkyloxy, aryloxyalkyloxy, arylthioalkyloxy, aryloxyalkylthio, arylthioalkylthio, arylalkylthio, aryloxyalkyl, arylthioalkyl, heterocyclic group, heterocyclic oxy, heterocyclic thio, heterocyclic alkyl, heterocyclic alkyloxy or heterocyclic alkylthio (provided that the alkyl present in these groups is straight-chain or branched alkyl having 1 to 4 carbon atoms) [a specific preferred example of the substituent is at least one group selected from the group consisting of:

a halogen atom, cyano, nitro, amino, hydroxyl, formyl, carboxyl, carbamoyl, thiocarbamoyl;

alkyl, alkoxy, alkylthio, alkylsulfinyl, or alkylsulfonyl, wherein said groups are straight-chain or branched groups having 1 to 6 carbon atoms;

straight-chain or branched alkenyl or alkenyloxy having 2 to 6 carbon atoms;

haloalkyl, haloalkoxy, haloalkylthio, haloalkylsulfinyl, or haloalkylsulfonyl, wherein said groups are straight-chain or branched groups having 1 to 6 carbon atoms that each have 1 to 13 halogen atoms which may be the same or different;

straight-chain or branched haloalkenyl having 2 to 6 carbon atoms or straight-chain or branched haloalkenyloxy having 2 to 6 carbon atoms, wherein said groups each have 1 to 13 halogen atoms which may be the same or different;

acylamino, N-acyl-N-alkylamino, alkylamino, dialkylamino, alkylcarbonyl, alkylcarbonyloxy, alkoxycarbonyl, alkylsulfonyloxy, hydroxyiminoalkyl or alkoxyiminoalkyl, wherein said groups each have straight-chain or branched alkyl having 1 to 6 carbon atoms;

alkylene, dioxyalkylene, or polyoxaalkylene, wherein said groups may be substituted by one, two or more substituents selected from the group consisting of a halogen atom, straight-chain or branched alkyl having 1 to 4 carbon atoms, and straight-chain or branched haloalkyl having 1 to 9 halogen atoms which may be the same or different, and are present as a chain which is substituted in its both ends at adjacent positions on the ring to form a ring; and

cycloalkyl having 3 to 6 carbon atoms or aryl]] (provided that the case where R_1 represents a hydrogen atom, A represents a bond or methylene, and R_3 represents phenyl or cyclohexyl, and the case where A represents an alkylene chain and R_3 represents a hydrogen atom are excluded).

[Claim 3]

The picolinamide derivative represented by formula (1) or a salt thereof according to claim 1 or 2, wherein

[A represents a bond, an ethylene chain or a methylene chain;

R_1 represents 4-methoxy or 6-methoxy;

R_2 is a hydrogen atom or benzyl;

R_3 is 4-phenoxyphenyl, 4-chlorophenyl, phenyl, 2-phenylcyclopropyl, cyclohexyl, 1-cyclohexenyl, 4-phenetoxyphenyl, 4-methylcyclohexyl, cycloheptyl, cyclooctyl, 4-(4-trifluoromethoxyphenoxy)phenyl, 4-(3-trifluoromethoxyphenoxy)phenyl, 3-methyl-4-(4-trifluoromethylphenoxy)phenyl, 3-methyl-4-(4-methylphenoxy)phenyl, or 3-methyl-4-(3-trifluorometnylphenoxy)phenyl].

[Claim 4]

A harmful organism control agent comprising the picolinamide derivative and/or a salt thereof according to any one of claims 1 to 3.

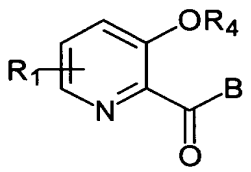
[Claim 5]

A harmful organism control agent comprising the picolinamide derivative and/or a salt thereof according to any one of claims 1 to 3 for exterminating plant pathogenic fungi.

[Claim 6]

A picolinic acid derivative represented by formula (2) or a salt thereof:

[Chem. 2]



(2)

wherein

[B represents hydroxyl, a halogen atom or alkoxy;

R_1 represents one, two or more groups, which may be the same or different, selected from the group consisting of alkoxy having 1 to 4 carbon atoms and haloalkoxy having 1 to 4 carbon atoms; and

R_4 represents a hydrogen atom, benzyl, alkyl having 1 to 4 carbon atoms or alkanoyl having 1 to 4 carbon atoms, in which the groups other than the hydrogen atom may be substituted (provided that the case where R_1 represents 4-methoxy with R_4 representing hydrogen or benzyl is excluded)].

[Claim 7]

A process for producing the picolinic acid derivative or a salt thereof according to claim 6, comprising the steps of:

oxidizing 2-hydroxymethylpyridine having a substituent in an inert solvent to form a 2-carboxyl compound; and

optionally removing the protective group by catalytic hydrogenation or hydrolysis (provided that the case where R_1 represents 4-methoxy and R_4 represents benzyl is excluded).

[Claim 8]

A process for producing the picolinic acid derivative or a salt thereof according to claim 6, comprising the steps of:

optionally introducing a protective group into 3-hydroxypicolinic acid to convert 3-hydroxypicolinic acid to an N-oxide compound;

successively subjecting the N-oxide compound to acylation and rearrangement to introduce acyloxy into the 6-positoin; and

optionally removing the protective group (provided that R_1 represents the alkoxy having 1 to 4 carbon atoms or haloalkoxy having 1 to 4 carbon atoms substituted at the 6-position).

[Claim 9]

A process for producing the picolinic acid derivative or a salt thereof according to claim 6 comprising the steps of:

optionally introducing a protective group into 3,4-disubstituted picolinic acid to convert 3,4-disubstituted picolinic acid to an N-oxide compound;

successively subjecting the N-oxide compound to acylation and rearrangement to introduce acyloxy into the 6- or 5-positoin; and

optionally removing the protective group (provided that R_1 represents alkoxy having 1 to 4 carbon atoms or haloalkoxy having 1 to 4 carbon atoms which may be the same or different and are substituted at the 4- and 5-positions or 4- and 6-positions).

[Detailed Description of the Invention]

[0001]

[Field of the Invention]

The present invention relates to a useful novel picolinamide derivative and a harmful organism control agent comprising said picolinamide derivative as an active component. The present invention also relates to a picolinic acid derivative as an intermediate indispensable for synthesizing a picolinamide derivative, and a process for producing the same.

[0002]

[Background Art]

Certain picolinamide derivatives are disclosed in Japanese Patent Laid-Open No. 242635/1995. This publication, however, does not disclose the use of the picolinamide derivatives as a harmful organism control agent. Further, the appearance of fungi resistance to existing various plant pathogenic fungi control agents has lead to an

ever-increasing demand for novel plant pathogenic fungi control agents.

[0003]

[Problems to be Solved by the Invention]

It is an object of the present invention to provide a novel picolinamide derivative useful for the control of harmful organisms for plant, and to provide a harmful organism control agent comprising the novel picolinamide derivative as an active component.

[0004]

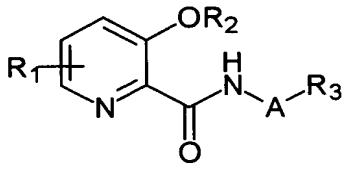
[Means for Solving the Problems]

The present inventors have found, as a result of devoted study to find more effective and safe fungi pathogenic control agent, that the picolinamide derivative represented by formula (1) has potent activity against harmful organisms for plant.

[0005]

That is, there is provided a picolinamide derivative represented by formula (1) or a salt thereof acceptable as an agricultural chemical and/or a harmful organism control agent comprising the picolinamide derivative as an active component:

[Chem. 3]



(1)

wherein

[A represents a bond or an optionally substituted alkylene chain;

R_1 represents a hydrogen atom or one, two or more groups, which may be the same or different, selected from the group consisting of, alkoxy, and haloalkoxy;

R_2 represents a hydrogen atom, benzyl, alkyl or alkanoyl, in which the groups other than the hydrogen atom may be substituted; and

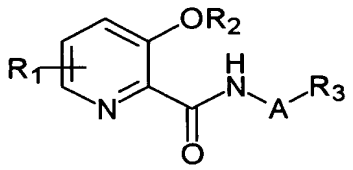
R_3 represents a hydrogen atom, cycloalkyl, cycloalkenyl, aryl or a heterocyclic

group, in which the groups other than the hydrogen atom may be substituted (provided that the case where R_1 represents a hydrogen atom, A represents a bond or a methylene chain, and R_3 represents phenyl or cyclohexyl, and the case where A represents an alkylene chain and R_3 represents a hydrogen atom are excluded)].

[0006]

The preferable compound represented by formula (1) is a picolinamide derivative or a salt thereof acceptable as an agricultural chemical and/or a harmful organism control agent comprising the picolinamide derivative as an active component:

[Chem. 4]



(1)

wherein

[A represents a bond or an alkylene chain having 1 to 12 carbon atoms;

R_1 represents a hydrogen atom or one, two or more groups, which may be the same or different, selected from the group consisting of alkoxy having 1 to 4 carbon atoms and haloalkoxy having 1 to 4 carbon atoms;

R_2 represents a hydrogen atom, benzyl having 1 to 4 carbon atoms, alkyl having 1 to 4 carbona toms, or alkanoyl having 1 to 4 carbon atoms, in which the groups other than the hydrogen atom may be substituted,

R_3 represents one, two or more groups, which may be the same or different, selected from optionally substituted cycloalkyl having 3 to 12 carbon atoms or cycloalkenyl having 3 to 12 carbon atoms [the substituent is one, two or more groups, which may be the same or different, selected from the group consisting of a halogen atom, cyano, nitro, carboxyl, hydroxyl, phenyl (phenyl may be optionally substituted by the substituent selected from one, two or more groups, which may be the same or different, selected from the group consisting of a halogen atom, cyano, nitro, amino, alkylamino, alkanoylamino, alkyl having 1 to 4 carbon atoms, haloalkyl having 1 to 4 carbon atoms, alkoxy having 1

to 4 carbon atoms, and haloalkoxy having 1 to 4 carbon atoms), alkyl having 1 to 4 carbon atoms, haloalkyl having 1 to 4 carbon atoms, or alkoxycabonyl having 1 to 4 carbon atoms];

optionally substituted monocyclic or polycyclic 3- to 12-membered aryl or 3- to 12-membered heterocyclic group [the substituent is one, two or more groups, which may be the same or different, selected from the group consisting of a halogen atom, cyano, nitro, amino, hydroxyl, formyl, carboxyl, carbamoyl, and thiocarbamoyl;

straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulfinyl, or alkylsulfonyl, having 1 to 6 carbon atoms;

straight-chain or branched alkenyl or alkenyloxy having 2 to 6 carbon atoms;

haloalkyl, haloalkoxy, haloalkylthio, haloalkylsulfinyl, or haloalkylsulfonyl, wherein said groups are straight-chain or branched groups having 1 to 6 carbon atoms that each have 1 to 13 halogen atoms which may be the same or different;

straight-chain or branched haloalkenyl, haloalkenyloxy having 2 to 6 carbon atoms that each have 1 to 13 halogen atoms which may be the same or different,

acylamino, N-acyl-N-alkylamino, alkylamino, dialkylamino, alkylcarbonyl, alkylcarbonyloxy, alkoxycarbonyl, alkylsulfonyloxy, hydroxyiminoalkyl, or alkoxyiminoalkyl, wherein said groups each have straight-chain or branched alkyl having 1 to 6 carbon atoms;

alkylene, dioxyalkylene, polyoxaalkylene, or cycloalkyl having 3 to 6 carbon atoms wherein said groups may be substituted by one, two or more substituents selected from the group consisting of a halogen atom, straight-chain or branched alkyl having 1 to 4 carbon atoms, and straight-chain or branched haloalkyl having 1 to 4 carbon atoms, which has 1 to 9 halogen atoms which may be the same or different, and are present as a chain which is substituted in its both ends at adjacent positions on the ring to form a ring]; and

aryl, aryloxy, arylthio, arylsulfinyl, arylsulfonyl, arylamino, arylalkyl, arylalkyloxy, aryloxyalkyloxy, arylthioalkyloxy, aryloxyalkylthio, arylthioalkylthio, arylalkylthio, aryloxyalkyl, arylthioalkyl, heterocyclic group, heterocyclic oxy, heterocyclic thio, heterocyclic alkyl, heterocyclic alkyloxy or heterocyclic alkylthio (provided that the

alkyl present in these groups is straight-chain or branched alkyl having 1 to 4 carbon atoms) [a specific preferred example of the substituent is a group selected from the group consisting of:

a halogen atom, cyano, nitro, amino, hydroxyl, formyl, carboxyl, carbamoyl, thiocarbamoyl;

alkyl, alkoxy, alkylthio, alkylsulfinyl, or alkylsulfonyl, wherein said groups are straight-chain or branched groups having 1 to 6 carbon atoms;

straight-chain or branched alkenyl or alkenyloxy having 2 to 6 carbon atoms;

haloalkyl, haloalkoxy, haloalkylthio, haloalkylsulfinyl, or haloalkylsulfonyl, wherein said groups are straight-chain or branched groups having 1 to 6 carbon atoms that each have 1 to 13 halogen atoms which may be the same or different;

straight-chain or branched haloalkenyl having 2 to 6 carbon atoms or straight-chain or branched haloalkenyloxy having 2 to 6 carbon atoms, wherein said groups each have 1 to 13 halogen atoms which may be the same or different;

acylamino, N-acyl-N-alkylamino, alkylamino, dialkylamino, alkylcarbonyl, alkylcarbonyloxy, alkoxycarbonyl, alkylsulfonyloxy, hydroxyiminoalkyl or alkoxyiminoalkyl, wherein said groups each have straight-chain or branched alkyl having 1 to 6 carbon atoms;

alkylene, dioxyalkylene, or polyoxaalkylene, wherein said groups may be substituted by one, two or more substituents selected from the group consisting of a halogen atom, straight-chain or branched alkyl having 1 to 4 carbon atoms, and straight-chain or branched haloalkyl having 1 to 4 carbon atoms, which has 1 to 9 halogen atoms which may be the same or different, and are present as a chain which is substituted in its both ends at adjacent positions on the ring to form a ring; and

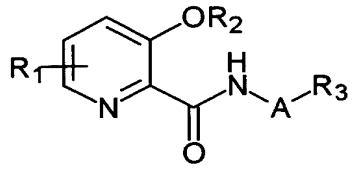
cycloalkyl having 3 to 6 carbon atoms or aryl]] (provided that the case where R_1 represents a hydrogen atom, A represents a bond or methylene, and R_3 represents phenyl or cyclohexyl, and the case where A represents an alkylene chain and R_3 represents a hydrogen atom are excluded).

[0007]

The specifically preferable compound represented by formula (1) is a picolinamide

derivative or a salt thereof acceptable as an agricultural chemical and/or a harmful organism control agent comprising the picolinamide derivative as an active component:

[Chem. 5]



(1)

wherein

[A represents a bond or ethylene,

R_1 represents 4-methoxy or 6-methoxy,

R_2 represents a hydrogen atom or benzyl,

R_3 represents 4-phenoxyphenyl, 4-chlorophenyl, phenyl, 2-phenylcyclopropyl, cyclohexyl, 1-cyclohexenyl, 4-phenetoxyphenyl, 4-methylcyclohexyl, cycloheptyl, cyclooctyl, 4-(4-trifluoromethoxyphenoxy)phenyl, 4-(3-trifluoromethoxyphenoxy)phenyl, 3-methyl-4-(4-trifluoromethylphenoxy)phenyl, 3-methyl-4-(4-methylphenoxy)phenyl, 3-methyl-4-(3-trifluoromethylphenoxy)phenyl].

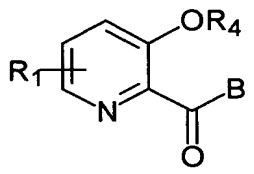
[0008]

Further, it is a second object of the present invention to provide the picolinic acid derivative represented by formula (2) and the process for producing the same.

[0009]

That is, there is provided a piclinic acid represented by formula (2) or an acid addition salt thereof:

[Chem. 6]



(2)

wherein

[B represents hydroxyl, a halogen atom or alkoxy;

R_1 represents one, two or more groups, which may be the same or different, selected from the group consisting of alkoxy having 1 to 4 carbon atoms and haloalkoxy having 1 to 4 carbon atoms; and

R_4 represents a hydrogen atom, benzy, alkyl having 1 to 4 carbon atoms or alkanoyl having 1 to 4 carbon atoms, in which the groups other than the hydrogen atom may be substituted (provided that the case where R_1 represents 4-methoxy is excluded)].

[0010]

A process for producing the picolinic acid derivative represented by formula (2) or a salt thereof, comprising the steps of: oxidizing 2-hydroxymethylpyridine having a substituent in an inert solvent to form a 2-carboxyl compound; and optionally removing the protective group by catalytic hydrogenation or hydrolysis (provided that the case where R_1 represents 4-methoxy and R_4 represents benzyl is excluded).

[0011]

A process for producing the picolinic acid derivative represented by formula (2) or a salt thereof, comprising the steps of: optionally introducing a protective group into 3-hydroxypicolinic acid to convert 3-hydroxypicolinic acid to an N-oxide compound; successively subjecting the N-oxide compound to acylation and rearrangement to introduce acyloxy into the 6-positoin; and optionally removing the protective group (provided that R_1 represents the alkoxy having 1 to 4 carbon atoms or haloalkoxy having 1 to 4 carbon atoms substituted at the 6-position).

[0012]

A process for producing the picolinic acid derivative represented by formula (2) or a salt thereof comprising the steps of: optionally introducing a protective group

into 3,4-disubstituted picolinic acid to convert 3,4-disubstituted picolinic acid to an N-oxide compound; successively subjecting the N-oxide compound to acylation and rearrangement to introduce acyloxy into the 6- or 5-positoin; and optionally removing the protective group (provided that R_1 represents alkoxy having 1 to 4 carbon atoms or haloalkoxy having 1 to 4 carbon atoms which may be the same or different and are substituted at the 4- and 5-positions or 4- and 6-positions).

[0013]

[Preferred Embodiments of the Invention]

The substituents defined in formula (1) representing the picolinic acid derivative according to the present invention will be described in detail.

[0014]

A represents a bond, methylene chain; 1,1- or 1,2-ethylene chain; 1,1-, 1,2-, 1,3- or 2,2-propylene chain; 1,1-, 1,2-, 1,3-, 1,4-, 2,2- or 2,3-butylene chain; 2-methyl-1,3-propylene chain; pentamethylene chain; hexamethylene chain; heptamethylene chain; octamethylene chain; nonamethylene chain; decamethylene chain; undecamethylene chain; or dodecamethylene chain.

[0015]

R_1 represents a hydrogen atom or one, two or more substituents, which may be the same or different, selected from the group consisting of methoxy, ethoxy, 1-propyloxy, isopropoxy, 1-butyloxy, 2-butyloxy, t-butyloxy, trifluoromethoxy, difluoromethoxy, fluorometoxy, difluorochloromethoxy and trifluoroethoxy.

[0016]

R_2 represents a hydrogen atom, benzyl, p-nitrobeyzyl, p-methoxybenzyl, methoxymethyl, methoxyethoxymethyl, isobutyril, acetyl, propionyl or pivaloyl.

[0017]

R_3 represents a hydrogen atom or monocyclic or polycyclic cycloalkyl or cycloalkenyl such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl, cyclooctyl, cyclononyl, cyclodecyl, cycloundecyl, cyclododecyl, cyclohexenyl, tetrahydronaphthyl, decahydronaphthyl, cyclododecatrienyl, indanyl, norbonyl, adamantyl, or cycloalkenyl which may be substituted by 1 to 6 substituents. The substituents may be

the same or different and are selected from the group consisting of a fluorine atom, a chlorine atom, a bromine atom, cyano, nitro, hydrochloride, carboxyl, methyl, ethyl n-propyl, isopropyl, phenyl, methoxy, ethoxy, methoxycarbonyl and ethoxycarbonyl.

[0018]

Or, aryl or heterocyclic group, which may be substituted by 1 to 6 substituents, such as phenyl, naphthyl, furyl, benzofuranyl, pyrrolyl, indolyl, thienyl, benzothienyl, oxazolyl, isoxazolyl, thiazolyl, isothiazolyl, oxadiazolyl, thiadiazolyl, pyridyl, quinolyl, pyrimidyl, pyridazinyl, pyrazinyl, oxiranyl, tetrahydrofuryl, perhydropiranyl, pyrrolidinyl, piperidinyl, homopiperidinyl or morpholinyl [specific examples of the substituents usable herein include: a fluorine atom, a chlorine atom, a bromine atom, cyano, nitro, amino, hydrochloride, formyl, carboxyl, carbamoyl, thiocarbamoyl, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, s-butyl, t-butyl, methoxy, ethoxy, n-propoxy, isopropoxy, methylthio, ethylthio, n-propylthio, isopropylthio, methylsulfinyl, methylsulfonyl, ethylsulfinyl, ethylsulfonyl, trifluoromethyl, trifluoroethyl, trifluoromethoxy, difluoromethoxy, difluorochloromethoxy, trifluoroethoxy, difluoromethylthio, difluorochloromethylthio, trifluoromethylthio, trifluoromethylfulfinyl, trifluoromethylsulfonyl, acetylamino, formylamino, N-formyl-N-methylamino, methylamino, ethylamino, n-propylamino, isopropylamino, dimethylamino, diethylamino, acetyl, propionyl, acetoxyl, methoxycarbonyl, ethoxycarbonyl, methylsulfonyloxy, ethylsulfonyloxy, hydroxyiminomethyl, hydroxyinimoethyl, methoxyiminomethyl, ethoxyiminomethyl, methoxyiminoethyl, and ethoxyiminoethyl].

[0019]

Or, trimethylene, tetramethylene, methylenedioxy, ethylenedioxy, or 1,4,7,10,13-pentaoxysatridecamethylene which may be substituted by one, two or more substituents, which may be the same or different, selected from the group consisting of a fluorine atom, a chlorine atom, methyl, trifluoromethyl, ethyl and n- or i-propyl, and is substituted in its both ends at adjacent positions on the ring to form a ring.

[0020]

Or, cyclopropyl, cyclobutyl, cyclopentyl or cyclohexyl.

[0021]

Or, phenyl, phenoxy, phenylalkyl, phenylthio, phenylsulfinyl, phenylsulfonyl, phenylcarbonyl, phenoxyalkyl, phenoxyalkyloxy, phenylthioalkyloxy, phenoxyalkylthio, phenylthioalkylthio, phenylthioalkyl, phenylalkyloxy, phenylalkyltho, pyridyl pyridyloxy, pyridylthio, anylino, morphonyl, piperidyl (provided that the alkyl chain is a straight-chain or branched alkyl chain having 1 to 4 carbon atoms) [specific examples of the substituents include a fluorine atom, a chlorine atom, a bromine atom, cyano, nitro, amino, formyl, carbamoyl, thiocarbamoyl, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, s-butyl, t-butyl, methoxy, ethoxy, n-propoxy, isopropoxy, methylthio, ethylthio, n-propylthio, isopropylthio, methylsulfinyl, methylsulfonyl, ethylsulfinyl, ethylsulfonyl, trifluoromethyl, trifluoroethyl, trifluoromethoxy, difluoromethoxy, difluorochloromethoxy, trifluoroethoxy, difluoromethylthio, difluorochloromethylthio, trifluoromethylthio, trifluoromethylsulfinyl, trifluoromethylsulfonyl, acetylamino, formylamino, N-formyl-N-methylamino, methylamino, ethylamino, n-propylamino, isopropylamino, dimethylamino, diethylamino, acetyl, propionyl, acetoxyl, methoxycarbonyl, ethoxycarbonyl, methylsulfonyloxy, ethylsulfonyloxy, methoxyiminomethyl, ethoxyiminomethyl, methoxyiminoethyl and ethoxyiminoethyl].

[0022]

Or, trimethylene, tetramethylene, methylenedioxy, ethylenedioxy, or 1,4,7,10,13-pentaoxysatridecamethylene which may be substituted by one, two or more substituents, which may be the same or different, selected from the group consisting of a fluorine atom, a chlorine atom, methyl, trifluoromethyl, ethyl and n- or i-propyl, and is substituted in its both ends at adjacent positions on the ring to form a ring.

[0023]

Or, cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl or phenyl.

[0024]

The substituents defined in formula (2) representing the picolinic acid derivative as an intermediate for the synthesis of drugs and agricultural chemicals will be described in detail.

[0025]

B represents hydroxyl, a cholorine atom, a bromine atom, methoxy, ethoxy,

methoxymethoxy, benzyloxy or 4-methoxybenzyloxy.

[0026]

R_1 represents one, two or more substituents, which may be the same or different, selected from the group consisting of methoxy, ethoxy, 1-propyloxy, isopropoxy, 1-butyloxy, 2-butyloxy, t-butyloxy, trifluoromethoxy, difluoromethoxy, fluoromethoxy, difluorochloromethoxy or trifluoroethoxy, specifically preferably, methoxy, ethoxy, trifluoromethoxy, difluoromethoxy, fluoromethoxy or difluorochloromethoxy.

[0027]

R_4 represents a hydrogen atom or optionally substituted benzyl, p-nitrobenzyl, p-methoxybenzyl, methoxymethyl, methoxyethoxymethyl or diphenylmethyl.

[0028]

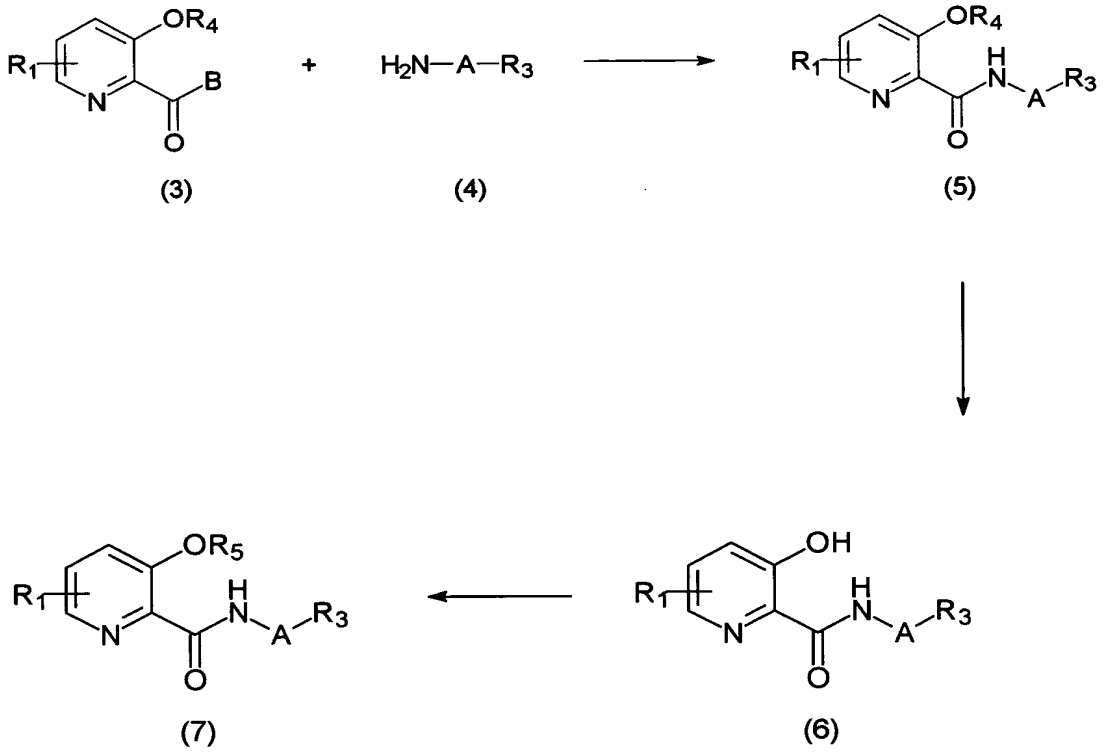
Next, the production process of a picolinamide derivative according to the present invention will be described in detail. However, it should be noted that the scope of the present invention is not limited by the following production process.

[0029]

The compound of formula (1) according to the present invention may be produced, for example, through a scheme 1 below, although the present invention is not limited to this scheme only.

[0030] Scheme 1

[Chem. 7]



[0031]

The compounds in scheme 1, A, B, R_1, R_3, and R_4 are as defined above. R_5 represents lower acyl, such as acetyl, propionyl or pivaloyl. The compounds of formulae (5), (6), and (7) are the compounds according to the present invention.

[0032]

According to this process, the picolinic acid derivative of formula (3) is reacted with an amine compound of formula (4) in the presence of a suitable condensation agent or an acid linking agent, or under aminolysis reaction conditions, in an inert solvent. Thereafter, when R_4 is a group other than a hydrogen atom, if necessary, the removal of R_4 and then optionally acylation are carried out to give picolinamide derivatives of formulae (5), (6) and (7).

[0033]

Condensation agents usable, in the case where B in formula (3) represents hydroxyl, include: acid halide formers, such as phosphorus trichloride, phosphorus

tribromide, phosphorus pentachloride, phsophorus oxychloride, and thionyl chloride; mixed acid anhydrides or acid halide of ethyl chloroformate and methanesulfonyl chloride; carbodiimides, such as N,N'-dicyclohexylcarbodiimide (DCC), 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide hydrochloride (WSCI·HCl); and other condensation agents, for example, N,N-carbonyldiimidazole, 2-ethoxy-N-ethoxycarbonyl-1,2-dihydroquinoline (EEDQ) and triphenylphosphine-carbon tetrachloride (complex).

[0034]

Alternatively, the picolinamide derivative may be produced by condensing 1-hydroxybenzotriazole or N-hydroxysuccinimid and a picolinic acid derivative with N,N'-dicyclohexylcarbodiimide to give an active ester compound which is then reacted with an amine compound.

[0035]

Further, when acid addition salts of a picolinic acid derivative and an amine compound are used addition of a base, such as triethylamine, can offer a smooth reaction.

[0036]

Solvents usable herein include: aromatic hydrocarbons, such as benzene, toluene and xylene; halogenated aromatic hydrocarbons, such as chlorobenzene and dichlorobenzene; aliphatic hydrocarbons, such as hexane, cyclohexane and petroleum ether; aliphatic halogenated hydrocarbons, such as dichloromethane, 1,2-chloroethane, chloroform and carbon tetrachloride; ethers, such as diethyl ether, diisopropyl ether, dioxane, tetrahydrofuran, ethylene glycol dimethyl ether and ethylene glycol diethyl ether; ketones, such as acetone, 2-butanone and methyl isobutyl ketone; nitriles, such as acetonitrile, propionitrile and benzonitrile; amides, such as N,N-dimethylformamide and hexamethylphosphoric triamide (HMPA); sulfoxides, such as dimethylsulfoxide; or mixtures thereof.

[0037]

The amount of reagents used in the reaction is not particularly limited. Preferably, however, based on one mol of the picolinic acid derivative represented by

formula (3), in general, the amine compound of formula (4) is used in an amount of 1.0 to 2.0 mol, preferably 1.0 to 1.3 mol, and the condensation agent is used in an amount of 1.0 to 5.0 mol, preferably 1.0 to 2.5 mol. The reaction temperature is not particularly limited. In general, however, the reaction temperature is in the range of -10℃ to the boiling temperature of the solvent used. The reaction time may vary depending upon concentration and temperature. In general, a reaction for 5 to 10 hr suffices for the production.

[0038]

Regarding the base added in the case where the acid addition salt of a picolinic acid derivative and an acid addition salt of an amine compound are used, a base may be used in an amount of 1.0 to 2.0 mol, preferably 1.0 to 1.3 mol, based on one mol of the acid addition salt of the picolinic acid derivative, and may be used in an amount of 1.0 to 2.0 mol, preferably 1.0 to 1.3 mol, based on one mol of the acid addition salt of the amine compound.

[0039]

Solvents usable, in the case where B in formula (3) represents a halogen atom, may be those described above. The acid linking agents usable herein include: alkali metal hydroxides or alkaline earth metal hydroxides, such as sodium hydroxide, potassium hydroxide, and calcium hydroxide; ammonium hydroxide; carbonates of alkali metals, such as sodium carbonate, potassium carbonate, sodium hydrogencarboante and potassium hydrogencarbonate; ammonium carbonate; acetates of alkali metals or alkaline earth metals, such as sodium acetate, potassium acetate and calcium acetate; ammonium acetate; hydrides of alkali metals or alkaline earth metals, such as sodium hydride, potassium hydride, and calcium hydride; and tertiary amines, such as trimethylamine, triethylamine, N,N-dimethylaniline, pyridine, 4-(dimethylamino)pyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) and diazabicycloundecene (DBU).

[0040]

The amount of the reagents used in the reaction is not particularly limited. Preferably, however, based on one mol of the acid halide of the 3-hydroxypicolinic acid

derivative, in general, the amine compound of formula (4) is used in an amount of 1.0 to 2.0 mol, preferably 1.0 to 1.3 mol, and the acid linking agent is used in an amount of 1.0 to 5.0 mol, preferably 1.0 to 2.5 mol. The reaction temperature is not particularly limited. In general, however, the reaction temperature is in the range of -10℃ to the boiling temperature of the solvent used. The reaction time may vary depending upon the concentration and temperature. In general, a reaction for 1 to 5 hr suffices for the production.

[0041]

Solvents usable, in the case where B in formula (3) represents alkoxy, may be those described above. The reaction may be carried out under conventional aminolysis conditions.

[0042]

The amount of the reagents used in the reaction is not particularly limited. Preferably, however, based on one mol of the alkoxy form of the 3-hydroxypicolinic acid derivative, in general, the amine compound of formula (4) is used in an amount of 1.0 to 10.0 mol, preferably 1.0 to 3.0 mol. The reaction temperature is not particularly limited. In general, however, the reaction temperature is in the range of -10°C to the boiling temperature of the solvent used. If necessary, the reaction is allowed to proceed under a pressure of 2 to 15 kbar. The reaction time may vary depending upon the concentration and temperature. In general, a reaction for 1 to 12 hr suffices for the production.

[0043]

If necessary, the picolinamide derivatives of formula (5) thus obtained, when R_4 represents a group other than a hydrogen atom, can be easily lead to a 3-hydroxy compound of formula (6) or an acid addition salt thereof by conventional methods.

[0044]

Methods usable herein are as follows. When R_4 represents optionally substituted benzyl, catalytic hydrogenation or acid hydrolysis is suitable. On the other hand, when R_4 represents methoxymethyl or methoxyethoxymethyl, acid hydrolysis is suitable. The 3-hydroxy compound thus obtained can be easily acylated by a conventional

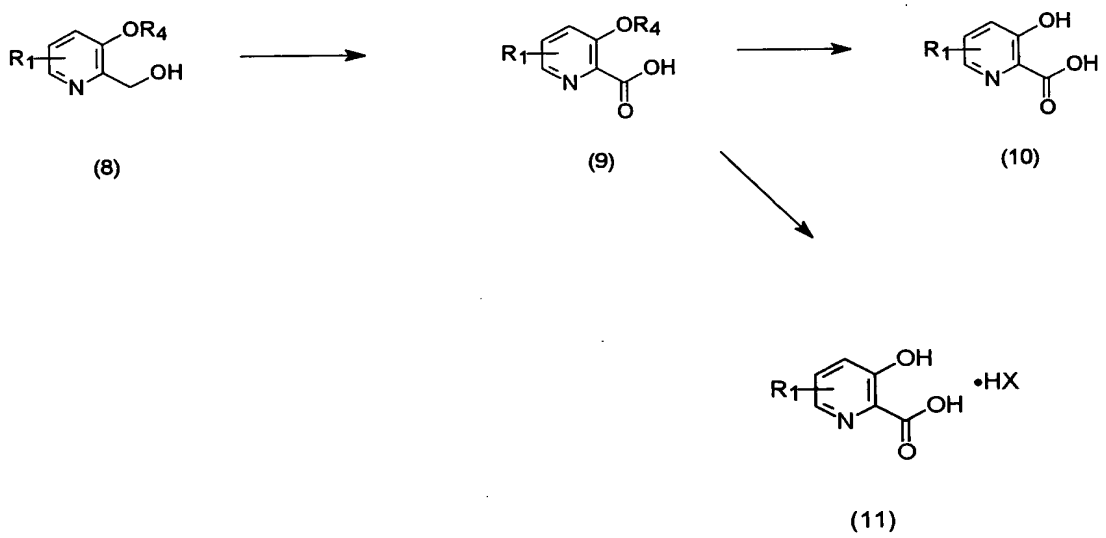
method to give a 3-acyloxy compound represented by formula (7). Solvents and acid linking agents usable herein may be those described above in connection with scheme 1. Acylation agents include acetic anhydride, propionic anhydride, acetyl chloride, acetyl bromide, propionyl chloride and pivaloyl chloride.

[0045]

The picolinic acid derivative represented by formula (2) according to the present invention is commercially available. The picolinic acid derivative of formula (2) according to the present invention may also be specifically produced by processes shown in the following schemes 2-1, 2-2 and 2-3. However, it should be noted that the scope of the present invention is not limited by these processes.

[0046] Scheme 2-1

[Chem. 8]



[0047]

In each picolinic acid derivative in scheme 2-1, R_1 represents one, two or more same or different alkoxys having 1 to 4 carbon atoms or haloalkoxys having 1 to 4 carbon atoms; R_4 represents a hydrogen atom, an optionally substituted benzyl, an optionally substituted alkyl having 1 to 4 carbon atoms or alkanoyl having 1 to 4 carbon atoms; and X represents a halogen atom, preferably a chlorine, bromine or iodine atom.

[0048]

According to the process shown in scheme 2-1, a substituted

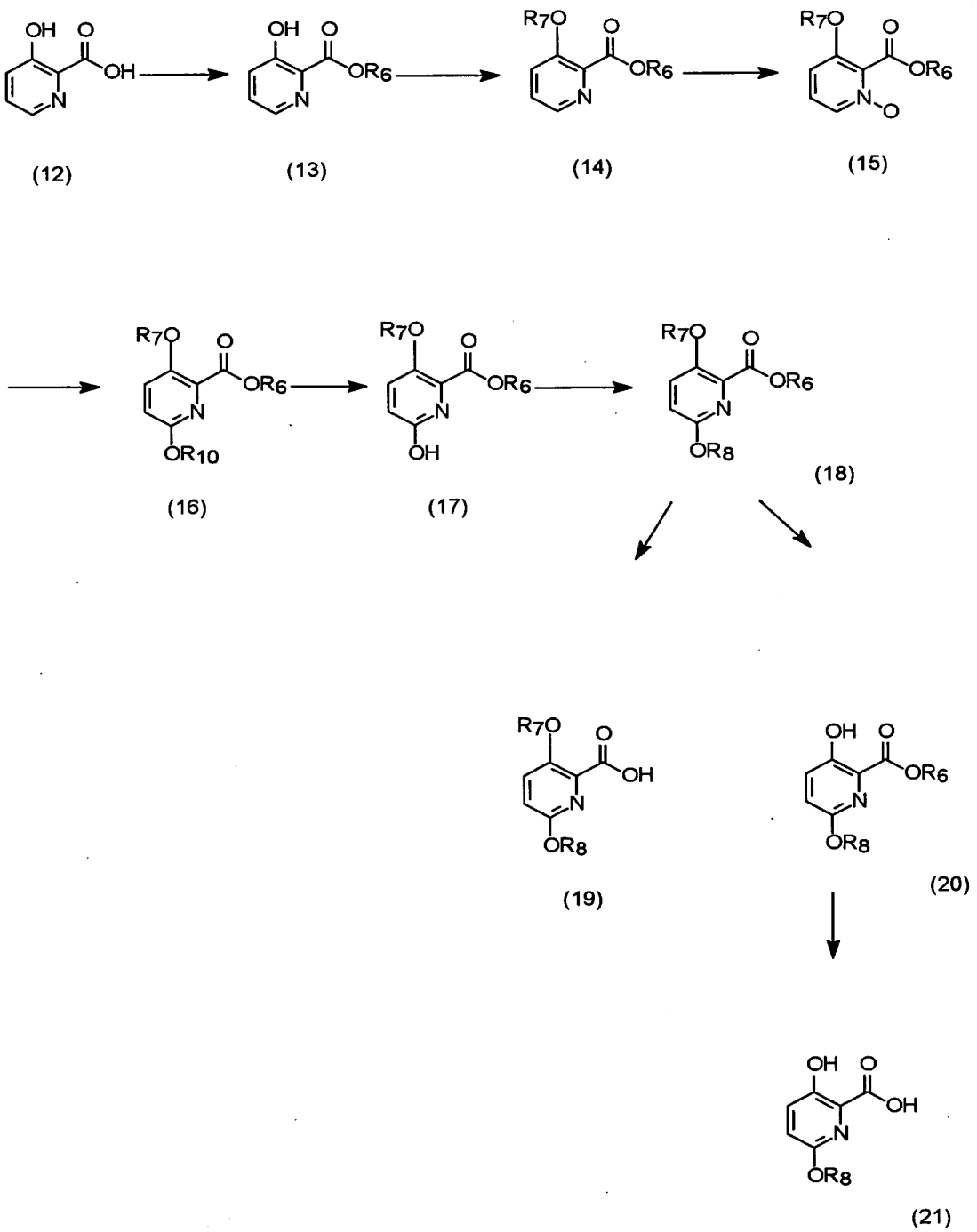
3-benzyloxy-2-hydroxymethylpyridine represented by formula (8), disclosed in EP 0208452 and EP 0304732, is oxidized in an inert solvent to give a substituted 3-benzyloxypicolinic acid represented by formula (9). Inert solvents include, for example, water. Oxidizing agents usable herein include, for example, potassium permanganate and sodium bichromate. The reaction temperature may vary depending upon the type of the reaction and the reagent and solvent used. In general, however, the reaction is carried out at about -20℃ to 100℃, preferably about 50 to 100℃. The reaction satisfactorily proceeds at a temperature of about 50 to 100℃ to give the title compound in high yield. Next, catalytic hydrogenation or acid hydrolysis is carried out to give a substituted 3-hydroxypicolinic acid of formula (10) or an acid addition salt thereof of formula (11). The catalytic hydrogenation or the acid hydrolysis can be easily carried out by a conventional method.

[0049]

Alternatively, 6-substituted 3-hydroxypicolinic acid or an acid addition salt thereof may be produced according to scheme 2-2.

[0050] Scheme 2-2

[Chem. 9]



[0051]

In each compound shown in scheme 2-2, R_6 represents alkyl having 1 to 8 carbon atoms; R_7 represents optionally substituted benzyl or optionally substituted alkyl

having 1 to 4 carbon atoms; R_8 represents alkoxy having 1 to 4 carbon atoms or haloalkoxy having 1 to 4 carbon atoms; and R_{10} represents formyl, acetyl, trichloroacetyl, trifluoroacetyl, chloroacetyl, propionyl, butyryl, isobutyryl, pivaloyl or phenoxyacetyl.

[0052]

Specifically, a commercially available 3-hydroxypicolinic acid represented by formula (12) is subjected to lower alkylation by a conventional esterification method. More specifically, the 3-hydroxypicolinic acid is treated with a corresponding lower alcohol in the presence of an acid catalyst, or alternatively is treated with a lower alkyl halide in the presence of a base in an inert solvent to give a 3-hydroxypicolinic ester of formula (13) in high yield. Here lower alkyl refers to alkyl having 1 to 8 carbon atoms, and suitable examples thereof include methyl, ethyl, n-propyl, isopropyl, n-butyl, and t-butyl. Acids usable as the acid catalyst include, for example, hydrogen chloride, sulfuric acid, and p-toluenesulfonic acid. The inert solvent is not particularly limited, and examples thereof include N,N-dimethylformamide, dimethylsulfoxide, acetonitrile, dioxane and tetrahydrofuran. Bases include: organic amines, such as triethylamine and pyridine; and inorganic bases, such as sodium carbonate and potassium carbonate. Lower alkyl halides include methyl iodide, ethyl iodide, ethyl bromide, 1-bromopropane, and 1-bromobutane. Alternatively, a simpler method may be used. Specifically, the 3-hydroxypicolinic acid may be treated with diazomethane or trimethylsilyldiazomethane in an inert solvent to give a methyl ester or may be treated with isobutene in the presence of an acid catalyst to give a t-butyl ester. The temperature used in these esterification reactions may vary depending upon the type of the reaction and the reagent and the solvent used. In general, however, the reaction temperature is about -20℃ to 100℃, preferably about 0 to 25℃. The reaction satisfactorily proceeds at this temperature to give the title compound in high yield.

[0053]

Next, a protective group is introduced into hydroxyl at the 3-position. The protective group is preferably removable under reduction conditions or acidic conditions. Examples of suitable protective groups include benzyl, p-methoxybenzyl, p-nitrobenzyl, methoxymethyl, methoxyethoxymethyl and diphenylmethyl. The compound can be

easily reacted with a corresponding halogenation reagent in an inert solvent in the presence of a base to convert the compound to the compound of formula (14). In the case of diphenylmethyl, the treatment with diphenyldiazomethane in an inert solvent is an optimal method. Examples of inert solvents include N,N-dimethylformamide, dimethylsulfoxide, acetonitrile, dioxane, tetrahydrofuran and acetone. Bases include sodium hydride and potassium carbonate. The halogen atom in the halogenation agent refers to chlorine, bromine or iodine. The reaction temperature is generally about 0 to 80℃, preferably about 25 to 50℃.

[0054]

The compound of formula (14) can be easily converted, by a conventional method involving the oxidation of nitrogen located within the pyridine ring, to an N-oxide compound of formula (15). The N-oxide compound of formula (15), when heated together with an acylation agent, is once converted to an N-acyloxy compound, and then causes a conventional thermal rearrangement reaction to give a 6-acyloxy compound of formula (16). Specific examples of suitable acyls include acyls having a small number of carbon atoms, such as formyl, acetyl, trichloroacetyl, trifluoroacetyl, propionyl, butyryl and isobutyryl. Among them, acetyl is most preferred. Acylating agents include corresponding carboxylic anhydride or acid chloride, and, in the case of acetylation, acetic anhydride is most preferred. Suitable reaction conditions are such that the reaction system is heated in the absence of a solvent or in the presence of an inert solvent (an inert solvent having a relatively high boiling point, such as toluene or xylene, being suitable) at 90 to 130℃. The 6-acyloxy compound of formula (16) may be deacylated under conventional basic conditions to give a 6-hydroxy compound of formula (17).

[0055]

Next, hydroxyl located at the 6-position of the 6-hydroxy compound of formula (17) is alkylated or haloalkylated to give a 6-alkoxy or 6-haloalkoxy compound of formula (18). In the case of methylation, diazomethane or trimethylsilyldiazomethane, which enables methylation under mild conditions, is suitable as an alkylation agent. In a general method, an alkylation agent, such as methyl iodide, dimethyl sulfate, methyl

p-toluenesulfonate, ethyl bromide, diethyl sulfate, 1-bromopropane, 1-bromobutane, or 1-bromopentane, or a haloalkylation agent, such as chloroiodomethane or iodotrifluoromethane, is used in an inert solvent (for example, N,N-dimethylformamide, dimethylsulfoxide, or acetone) in the presence of a base (for example, sodium hydride, t-butoxypotassium or potassium carbonate). The reaction temperature is in the range of about 0 to 80℃, preferably in the range of about 25 to 60℃.

[0056]

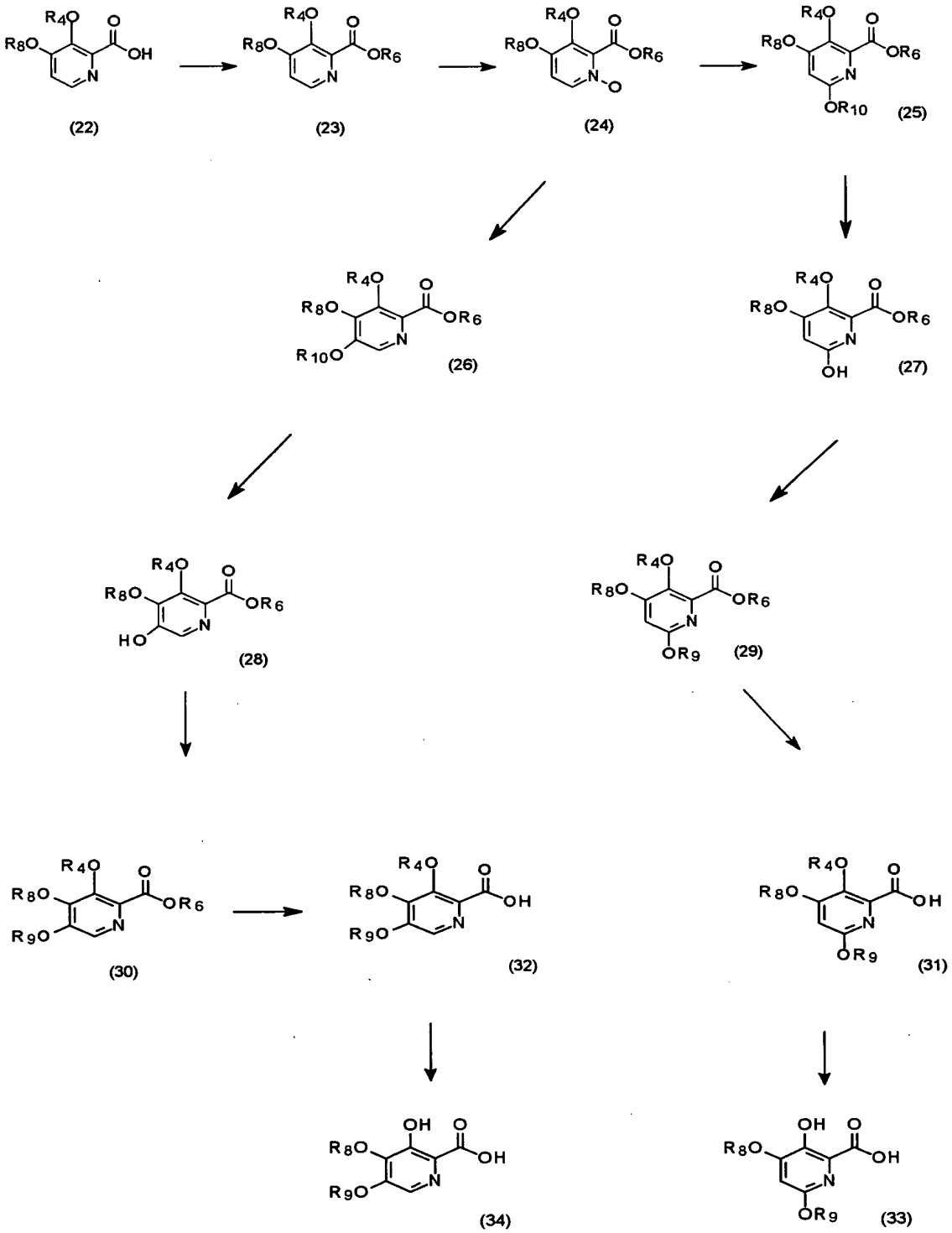
Finally, the removal of the protective group for hydroxyl at the 3-position and the deesterification of the carobxyl at the 2-positon can be easily carried out by conventional methods. Thus, a deesterification product of formula (19), a compound of formula (20) wherein the protective group at the 3-positon has been removed, and a 3-hydroxy-6-substituted picolinic acid of formula (21) or an acid addition salt thereof can be obtained.

[0057]

Further, 4,6-Disubstituted 3-hydroxypicolinic acid, 4,5-disubstituted 3-hydroxypicolinic acid, or an acid addition salt thereof may also be produced according to scheme 2-3.

[0058] Scheme 2-3

[Chem. 10]



[0059]

In the compounds in scheme 2-3, the substituents R_4, R_6, R_8 and R_{10} are as defined above; and R_9 represents alkoxy having 1 to 4 carbon atoms or haloalkoxy having 1 to 4 carbon atoms.

[0060]

That is, parts of the products in step 2-1, compound of formula (22) are provided as a starting compounds, and are esterified and oxidized in the similar manner as used in connection with 3-hydroxy-6-substituted picolinic acid to give a 4-substituted-3-hydroxypicolinic ester of formula (23) which is then converted to an N-oxide compound of formula (24). The acylation is then carried out in the similar manner as in the case of 3-hydroxy-6-substituted picolinic acid, and is subjected to a rearrangement reaction. In this case, both compounds of formula (25), wherein acyloxy has been rearranged to the 6-positoin, and a compound of formula (26), wherein acyloxy has been rearranged to the 5-position, are produced. These compounds can be easily separated by silica gel chromatography. In the similar manner as used above in connection with 3-hydroxy-6-substituted picolinic acid, these rearrangement products can be deacylated to give compounds of formulae (27 and 28), and, subsequently, alkylation or haloalkylation of hydroxyl at the 6-position or 5-position are carried out to give a 4,6-disubstituted compound of formula (29) and a 4,5-disubstituted compound of formula (30).

[0061]

Next, deesterification can be carried out by a conventional method to give a 4,6-disubstituted-3-benzyloxypicolinic acid of formula (31) and a 4,5-disubstituted-3-benzyloxypicolinic acid of formula (32) or an acid addition salt thereof. Thereafter, if necessary, the removal of the protective group for hydroxyl at the 3-position can be carried out by a conventional method to give a 4,6-disubstituted-3-hydroxypicolinic acid of formula (33) and a 4,5-disubstituted-3-hydroxypicolinic acid of formula (34) or an acid addition salt thereof.

[0062]

Further, the picolinic acid derivatives of formula (2), except for the case where R_1 represents hydrogen or 4-methoxy, are novel compounds. Further, the picolinamide

derivatives of formulae (5) to (7) have high harmful organism control activity and thus are very useful as an intermediate for the synthesis of drugs and agricultural chemicals.

[0063]

Further, the amines of formula (4) are commercially available or may be produced by a conventional process.

[0064]

The reaction mixture containing the contemplated compound of the present invention can be purified by extraction, concentration, filtration, chromatography, recrystallization and other conventional means.

[0065]

When using the compound according to the present invention as an active component of a bactricide, the compound may be used as such without being added with any additive. However, the compound is usually mixed with a solid carrier, a liquid carrier, a gaseous carrier, and a bait, and, if necessary, a surfactant and other additives for preparations are added thereto to formulate the control agent into oil solutions, emulsifiable concentrates, wettable powder, floables, granules, dust, aerosols and sprays in use. In any of these products, preferably, the compound of the present invention is generally contained in an amount of about 0.01 to 95% by weight. More preferably, in the case of oils, the compound is contained in an amount of about 0.5 to 5% by weight; in the case of emulsifiable concentrates, is contained in an amount of about 1 to 50% by weight; in the case of wettable powders and floables, is contained in an amount of about 1 to 90% by weight; in the case of granules, is contained in an amount of about 0.5 to 25% by weight; in the case of dust, is contained in an amount of about 0.3 to 25% by weight; and in the case of aerosols, is contained in an amount of about 0.1 to 5% by weight.

[0066]

The compounds thus prepared may be applied to, for example, foliage application, submerged application, soil treatment, nursery cabinet treatment, fumigation, seed sterilization, facility sterilization and equipment sterilization. Needless to say, the compounds are also effective in other forms of application by those skilled in the art.

[0067]

Solid carrier usable in the formulation include, for example, fine powders or particulates of clays (for example, kaolin clay, diatomaceous earth, synthetic hydrous silicon oxide, bentonite, fubasami clay, acid clay), talcs, ceramics and other inorganic minerals (for example, celite, quartz, sulfur, activated carbon, calcium carbonate and hydrous silica), chemical fertilizers (for example, ammonium sulfate, ammonium phosphate, ammonium nitrate, urea and ammonium chloride). Liquid carriers include, for example, water, alcohols (for example, methanol and ethanol), ketones (for example, acetone and methyl ethyl ketone), aromatic hydrocarbons (for example, benzene, toluene, xylene, ethylbenzene and methylnaphthalene), aliphatic hydrocarbons (for example, hexane, cyclohexane, kerosene and gas oil), esters (for example, ethyl acetate and butyl acetate), nitriles (for example, acetonitrile and isobutyronitrile), ethers (for example, diisopropyl ether and dioxane), acid amides (for example, N,N-dimethylformamide and N,N-dimethylacetamide), halogenated hydrocarbons (for example, dichloromethane, trichloroethane and carbon tetrachloride), dimethyl sulfoxide and vegetable oils such as soybean oil and cotton seed oil. Gaseous carriers, that is, propellants, include, for example, butane gas, LPG (liquefied petroleum gas), dimethyl ether and carbon dioxide.

[0068]

Additives preparations include, for example, fixing agents or dispersants, such as casein, gelatin, polysaccharides (for example, starch powder, gum arabic, cellulose derivatives and arginic acid), lignin derivatives, bentonite, saccharides, synthetic water-soluble polymers (for example, polyvinyl alcohol, polyvinylpyrrolidone, and polyacrylic acids), for example, PAP (for example, acidic isopropyl phosphate), BHT (for example, 2,6-di-tert-butyl-4-methylphenol), BHA (for example, a mixture of 2-tert-butyl-4-methoxyphenol with 3-tert-butyl-4-methoxyphenol), vegetable oils, mineral oils, surfactants, stabilizers such as fatty acids (for example, stearic acid), their esters or salts.

[0069]

In use, the compounds thus prepared may be used either as such or after dilution with water. Alternatively, the compounds may be used in combination with or

as a mixture with other bactericides, nematicides, miticides, herbicides, growth-regulating substances of plants, or synergists.

[0070]

Surfactants include, for example, alkylsulfonic esters, alkylsulfonic acid salts, alkylarylsulfonic acid salts, alkyl aryl ethers, and polyoxyethylenation products thereof, polyethylene glycol ethers, polyhydric alcohol esters and sugar alcohol derivatives.

[0071]

When using the compound of the present invention as a bactericide for agriculture and gardening, the application rate is generally about 0.1 to 100 g per 10 ares, in terms of the active component. When the emulsifiable concentrate, the wettable powder, or the floables is used after dilution with water, the application concentration is generally about 0.1 to 1000 ppm, and the granules and the dust are preferably applied as such without dilution. The application rate and application concentration of the compound may be increased or decreased regardless of the above described ranges depending upon type, application season, application sites, application methods, type of diseases, and level of damage.

[0072]

The compound of the present invention is useful for various diseases harmful to agriculture and gardening, for examples, various diseases of vegetables, fruit trees, paddy rice, or garden plants, and is very useful for plant diseases caused by representative plant pathogenic fungi belonging to deuteromyces, ascomycontina, and basidiomycetes. In particular, the compound of the present invention has significant control effect against plant diseases, such as rice blast, cucumber anthracnose, powdery mildew of cucumber, and wheat leaf rust.

[0073]

[Examples]

The following examples, preparation examples, and evaluation test examples further illustrate the present invention, but should not be construed as limiting the scope of the present invention. It should be noted that the examples of the present invention are illustrative only and conventional means may be applied according to the properties

of the picolinic acid derivatives clarified by the present invention to perform synthesis, extraction, purification, and utilization.

[0074]

Production Example 1

3-Hydroxy-4'-phenoxypicolinanilide:

3-Hydroxypicolinic acid 1.39 g (10.0 mmol) and 1.95 g (12.0 mmol) of carbonyldiimidazole were mixed into anhydrous N,N-dimethylformamide (hereinafter referred to as "DMF") to prepare a suspension (30 ml). An anhydrous DMF solution (25 ml) of 1.85 g (10.0 mmol) of 4-phenoxyaniline was added dropwise to this suspension, and the reaction was allowed to proceed at room temperature overnight. Water (50 ml) was added to the reaction mixture, followed by extraction with ethyl acetate. The organic layer was dried over anhydrous sodium sulfate, and the dried organic layer was concentrated under the reduced pressure. The residue was purified by column chromatography on silica gel (ethyl acetate-n-hexane) to give 1.24 g (yield 41%) of the title compound.

[0075]

The procedure of Production Example 1 was repeated to give the compounds of the following Production Examples 2 to 7.

[0076]

Production Example 2

3-Hydroxy-4'-benzylpicolinanilide

[0077]

Production Example 3

3-Hydroxy-4'-(2,6-di-sec-butylphenoxy)picolinanilide

[0078]

Production Example 4

3-Hydroxy-4'-(4-t-butylphenoxy)picolinanilide

[0079]

Production Example 5

3-Hydroxy-4'-(2,4-di-t-butylphenoxy)picolinanilide

[0080]

Production Example 6

3-Hydroxy-4'-(3-trifluoromethylphenoxy)picolinanilide

[0081]

Production Example 7

3-Hydroxy-N-cyclohexylpicolinamide

[0082]

Production Example 8

3-Benzyloxy-4-methoxy-4'-phenoxypicolinanilide:

3-Benzyloxy-4-methoxypicolinic acid 0.65 g (2.5 mmol) and 0.50 g (3.0 mmol) of carbonyldiimidazole were mixed into anhydrous DMF to prepare a suspension (8 ml). An anhydrous DMF solution (2 ml) of 0.56 g (3.0 mmol) of 4-phenoxyaniline was added dropwise to this suspension, and a reaction was allowed to proceed at room temperature overnight. Water 10 ml was added to the reaction mixture, followed by extraction with ethyl acetate. The organic layer was dried over anhydrous sodium sulfate, and the dried organic layer was concentrated under the reduced pressure. The residue was purified by column chromatography on silica gel (ethyl acetate-n-hexane) to give 0.76 g (yield 71%) of the title compound.

[0083]

Production Example 9

3-Hydroxy-4-methoxy-4'-phenoxypicolinanilide:

3-Benzyloxy-4-methoxy-4'-phenoxypicolinanilide 0.64 g (1.5 mmol) was mixed with ethanol (4 ml) to prepare a suspension. To this suspension was added 64 mg of 10% palladium-carbon. The mixture was subjected to catalytic reduction under atmospheric conditions overnight. The reaction solution was filtered, and the filtrate was concentrated under the reduced pressure. The residue was dissolved in a water-methanol mixed solution, and was recrystallized to give 0.41 g (yield 81%) of the title compound.

[0084]

The procedure of Production Examples 8 and 9 was repeated to prepare the compounds of the following Production Examples 10 to 24.

[0085]

Production Example 10

3-Hydroxy-4-methoxy-4'-(4-t-butylphenoxy)picolinanilide

[0086]

Production Example 11

3-Hydroxy-4-methoxy-3'-phenoxypicolinanilide

[0087]

Production Example 12

3-Hydroxy-4-methoxy-2'-phenoxypicolinanilide

[0088]

Production Example 13

3-Hydroxy-4-methoxy-4'-benzylpicolinanilide

[0089]

Production Example 14

3-Hydroxy-4-methoxy-4'-phenylthiopicolinanilide

[0090]

Production Example 15

3-Hydroxy-4-methoxy-4'-(4'-methoxyphenoxy)picolinanilide

[0091]

Production Example 16

3-Hydroxy-4-methoxy-3'-trifluoromethyl-4'-(4-trifluoromethylphenoxy)picolinanilide

[0092]

Production Example 17

3-Hydroxy-4-methoxy-4'-(4-phenylphenoxy)picolinanilide

[0093]

Production Example 18

3-Hydroxy-4-methoxy-4'-(4-methylphenoxy)picolinanilide

[0094]

Production Example 19

3-Hydroxy-4-methoxy-4'-(4-methylphenoxy)-3'-trifluoromethylpicolinanilide

[0095]

Production Example 20

3-Hydroxy-4-methoxy-2'-methoxy-4'-phenoxypicolinanilide

[0096]

Production Example 21

3-Hydroxy-4-methoxy-3'-chloro-4'-phenoxypicolinanilide

[0097]

Production Example 22

3-Hydroxy-4-methoxy-4'-phenoxy-3'-trifluoromethylpicolinanilide

[0098]

Production Example 23

3-Hydroxy-4-methoxy-3'-methyl-4'-phenoxypicolinanilide

[0099]

Production Example 24

3-Hydroxy-4-methoxy-2'-methoxy-4'-(4-methylphenoxy)picolinanilide

[0100]

Production Example 25

3-Hydroxy-4-methoxy-4'-(2,4-di-t-butylphenoxy)picolinanilide:

3-Hydroxy-4-methoxypicolinic acid 0.20 g (1.18 mmol) and 0.23 g (1.42 mmol) of carbonyldiimidazole were mixed into DMF to prepare a suspension (5 ml). An anhydrous DMF solution 1 ml of 0.35 g (1.18 mmol) of 4-(2,4-di-t-butylphenoxy)-aniline was added dropwise to this suspension, and a reaction was allowed to proceed at room temperature for 2 days. Water 5 ml was added to the reaction mixture, followed by extraction with ethyl acetate. The organic layer was dried over anhydrous sodium sulfate, and the dried organic layer was concentrated under the reduced pressure. The residue was purified by column chromatography on silica gel (ethyl acetate-n-hexane) to give 0.19 g (yield 36%) of the title compound.

[0101]

Production Example 26

3-Hydroxy-4-methoxy-4'-(3-trifluoromethylphenoxy)picolinanilide:

4-(3-Trifluoromethylphenoxy)aniline 0.15 g (0.59 mmol) and 0.15 g (0.72 mmol) of dicyclohexylcarbodiimide were added to a suspension (5 ml) of 0.10 g (0.59 mmol) of 3-hydroxy-4-methoxypicolinic acid in anhydrous pyridine, and a reaction was allowed to proceed at 90°C for 3 hr. The reaction mixture was cooled, and was then filtered. The filtrate was concentrated under the reduced pressure. To the concentrate was added 5 ml of 0.5 M hydrochloric acid. The mixture was vigorously stirred. The resultant precipitate was collected by filtration, was washed with 5 ml of cold water, and was then purified by column chromatography on silica gel (ethyl acetate-n-hexane) to give 0.06 g (yield 25%) of the title compound.

[0102]

The procedure of Production Example 26 was repeated to prepare the compounds of the following Production Examples 27 and 28.

[0103]

Production Example 27

3-Hydroxy-4-methoxy-4'-(3,5-di-t-butylphenoxy)picolinanilide

[0104]

Production Example 28

3-Hydroxy-4-methoxy-3'-chloro-4'-(4-chlorophenoxy)picolinanilide

[0105]

Production Example 29

3-Hydroxy-4-methoxy-4'-(4-methoxyphenoxy)-3'-methylpicolinanilide:

4-(4-Methoxyphenoxy)aniline 0.23 g (1.00 mmol), 0.26 g (1.00 mmol) of 3-benzyloxy-4-methoxypicolinic acid, and 0.20 g (1.50 mmol) of 1-hydroxybenzotriazole were mixed into chloroform to prepare a suspension (8 ml). WSCI·HCl 0.29 g (1.5 mmol), a chloroform solution (4 ml), and 0.15 g (1.5 mmol) of triethylamine were added dropwise at -20℃ to this suspension. Thereafter, a reaction was allowed to proceed at room temperature overnight. The reaction mixture was concentrated under the reduced pressure. The concentrate was dissolved in chloroform. The solution was washed with saturated brine, and was then dried over anhydrous sodium sulfate. The dried solution was concentrated and dried under the reduced pressure. The residue was

purified by column chromatography on silica gel (chloroform) to give 0.41 g of 3-benzyloxy-4-methoxy-4'-(4-methoxyphenoxy)-3'-methylpicolylanilide. This product was suspended in 5 ml of ethanol. To the suspension was added 30 mg of 10% palladium-carbon. The mixture was subjected to catalytic reduction under atmospheric conditions overnight. The reaction solution was filtered, and the filtrate was concentrated under the reduced pressure. The residue was then purified by column chromatography on silica gel (chloroform) to give 0.21 g (yield 55%) of the title compound.

[0106]

The procedure of Production Example 29 was repeated to prepare the compounds of the following Production Examples 30 to 38.

[0107]

Production Example 30

3-Hydroxy-4-methoxy-N-(1-(1-naphthyl)ethyl)picolinamide

[0108]

Production Example 31

3-Hydroxy-4-methoxy-3'-chloro-4'-(4-methoxyphenoxy)picolinanilide

[0109]

Production example 32

3-Hydroxy-4-methoxy-3'-chloro-4'-(4-methylphenoxy)picolinanilide

[0110]

Production Example 33

3-Hydroxy-4-methoxy-3'-methyl-4'-(4-methylphenoxy)picolinanilide

[0111]

Production Example 34

3-Hydroxy-4-methoxy-4'-(4-trifluoromethoxyphenoxy)picolinanilide

[0112]

Production Example 35

3-Hydroxy-4-methoxy-4'-(3-trifluoromethoxyphenoxy)picolinanilide

[0113]

Production Example 36

3-Hydroxy-4-methoxy-4'-(4-methylphenoxy)-2-trifluoromethylpicolinanilide

[0114]

Production Example 37

3-Hydroxy-4-methoxy-2',4'-di(4-methylphenoxy)picolinanilide

[0115]

Production Example 38

3-Hydroxy-4-methoxy-3',5'-di-t-butylpicolinanilide

[0116]

Production Example 39

3-Hydroxy-4-methoxy-4'-benzyloxypicolinanilide:

4-Benzyloxyaniline hydrochloride 0.21 g (0.87 mmol), 0.15 g (0.73 mmol) of 3-hydroxy-4-methoxypicolinic acid hydrochloride, 0.15 g (1.10 mmol) of 1-hydroxybenzotriazole, and 0.16 g (1.10 mmol) of triethylamine were mixed into chloroform to prepare a suspension (2 ml). A chloroform solution (2 ml) of WSCI·HCl 0.21 g (10 mmol) and 0.11 g (1.10 mmol) of triethylamine were added dropwise at -20℃ to this suspension, and a reaction was then allowed to proceed at room temperature overnight. The reaction mixture was concentrated under the reduced pressure. The concentrate was redissolved in chloroform. The solution was washed with saturated brine, and was then dried over anhydrous sodium sulfate. The dried solution was concentrated under the reduced pressure. The residue was purified by column chromatography on silica gel (chloroform) to give 0.15 g (yield 59%) of the title compound.

[0117]

The procedure of Production Example 39 was repeated to prepare the compounds of the following Production Examples 40 to 88.

[0118]

Production Example 40

3-Hydroxy-4-methoxy-3'-benzyloxypicolinanilide

[0119]

Production Example 41

3-Hydroxy-4-methoxy-3'-(3-pyridyl)picolinanilide

[0120]

Production Example 42

3-Hydroxy-4-methoxy-N-cyclododecylpicolinamide

[0121]

Production Example 43

3-Hydroxy-4-methoxy-N-cyclooctylpicolinamide

[0122]

Production Example 44

3-Hydroxy-4-methoxy-4'-(phenylamino)picolinanilide

[0123]

Production Example 45

3-Hydroxy-4-methoxy-N-(1-adamantyl)picolinamide

[0124]

Production Example 46

3-Hydroxy-4-methoxy-4'-(4-morpholinyl)picolinanilide

[0125]

Production Example 47

3-Hydroxy-4-methoxy-N-(1-adamantanemethyl)picolinamide

[0126]

Production Example 48

3-Hydroxy-4-methoxy-3'-methyl-4'-(3-trifluoromethylphenoxy)picolinanilide

[0127]

Production Example 49

3-Hydroxy-4-methoxy-4'-cyclohexylpicolinanilide

[0128]

Production Example 50

3-Hydroxy-4-methoxy-N-(4-benzo-15-crown-5-yl)picolinamide

[0129]

Production Example 51

3-Hydroxy-4-methoxy-(3',4'-ethylenedioxy)picolinanilide

[0130]

Production Example 52

3-Hydroxy-4-methoxy-N-(1-benzylpiperidin-4-yl)picolinamide

[0131]

Production Example 53

3-Hydroxy-4-methoxy-N-(2-(1-cyclohexenyl)ethyl)picolinamide

[0132]

Production Example 54

3-Hydroxy-4-methoxy-4'-(4-nitrophenoxy)picolinanilide

[0133]

Production Example 55

3-Hydroxy-4-methoxy-2',6'-dimethyl-4'-phenoxypicolinanilide

[0134]

Production Example 56

3-Hydroxy-4-methoxy-N-(2-trans-phenylcyclopropyl)picolinamide

[0135]

Production Example 57

3-Hydroxy-4-methoxy-N-cycloheptylpicolinamide

[0136]

Production Example 58

3-Hydroxy-4-methoxy-4'-(4-N-isopropylaminophenoxy)picolinanilide

[0137]

Production Example 59

3-Hydroxy-4-methoxy-N-cyclohexylpicolinamide

[0138]

Production Example 60

3-Hydroxy-4-methoxypicolinanilide

[0139]

Production Example 61

3-Hydroxy-4-methoxy-4'-chloropicolinanilide

[0140]

Production Example 62

3-Hydroxy-4-methoxy-4'-(4-aminophenoxy)picolinanilide

[0141]

Production Example 63

3-Hydroxy-4-methoxy-N-(2-cyclohexylethyl)picolinamide

[0142]

Production Example 64

3-Hydroxy-4-methoxy-4'-benzoylpicolinanilide

[0143]

Production Example 65

3-Hydroxy-4-methoxy-N-(1-indanyl)picolinamide

[0144]

Production Example 66

3-Hydroxy-4-methoxy-N-(1,2,3,4-tetrahydronaphtho-1-yl)picolinamide

[0145]

Production Example 67

3-Hydroxy-4-methoxy-N-benzylpicolinamide

[0146]

Production Example 68

3-Hydroxy-4-methoxy-N-phenetylpicolinamide

[0147]

Production Example 69

3-Hydroxy-4-methoxy-N-(1-phenylethyl)picolinamide

[0148]

Production Example 70

3-Hydroxy-4-methoxy-N-(1-methyl-1-phenylethyl)picolinamide

[0149]

Production Example 71

3-Hydroxy-4-methoxy-N-(4-phenoxybenzyl)picolinamide

[0150]

Production Example 72

3-Hydroxy-4-methoxy-4'-(4-phenetyloxy)picolinanilide

[0151]

Production Example 73

3-Hydroxy-4-methoxy-4'-(4-isobutyrylpiperazin-1-yl)picolinanilide

[0152]

Production Example 74

3-Hydroxy-4-methoxy-N-(1-homopiperidyl)picolinamide

[0153]

Production Example 75

3-Hydroxy-4-methoxy-N-(cyclohexylmethyl)picolinamide

[0154]

Production Example 76

3-Hydroxy-4-methoxy-N-(2-trans-methylcyclohexyl)picolinamide

[0155]

Production Example 77

3-Hydroxy-4-methoxy-N-(2-cis-methylcyclohexyl)picolinamide

[0156]

Production Example 78

3-Hydroxy-4-methoxy-N-(4-methylcyclohexyl)picolinamide

[0157]

Production Example 79

3-Hydroxy-4-methoxy-N-cyclopentylpicolinamide

[0158]

Production Example 80

3-Hydroxy-4-methoxy-N-cyclopropylpicolinamide

[0159]

Production Example 81

3-Hydroxy-4-methoxy-N-cyclobutylpicolinamide

[0160]

Production Example 82

3-Hydroxy-4-methoxy-N-(2-butyl)picolinamide

[0161]

Production Example 83

3-Hydroxy-4-methoxy-N-(n-hexyl)picolinamide

[0162]

Production Example 84

3-Hydroxy-4-methoxy-N-(4-hydroxycyclohexyl)picolinamide

[0163]

Production Example 85

3-Hydroxy-4-methoxy-N-(2-hydroxycyclohexyl)picolinamide

[0164]

Production Example 86

3-Hydroxy-4-methoxy-N-(n-octyl)picolinamide

[0165]

Production Example 87

3-Hydroxy-4-methoxy-N-(n-heptyl)picolinamide

[0166]

Production Example 88

3-Hydroxy-4-methoxy-N-(3,3-dimethylbutyl)picolinamide

[0167]

Production Example 89

3-Benzyloxy-6-methoxy-4'-phenoxypicolinanilide:

The procedure of Production Example 29 was repeated, except that 3-benzyloxy-4-methoxypicolinic acid was changed to 3-benzyloxy-6-methoxypicolinic acid. Thus, the title compound was prepared (yield 57%).

[0168]

Production Example 90

3-Hydroxy-6-methoxy-4'-phenoxypicolinanilide:

The procedure of Production Example 29 was repeated, except that 3-benzyloxy-6-methoxy-4'-phenoxypicolinanilide was subjected to catalytic reduction to give the title compound (yield 83%).

[0169]

The procedure of Production Examples 89 and 90 was repeated to prepare the compounds of the following Production Examples 91 to 93.

[0170]

Production Example 91

3-Hydroxy-6-methoxy-N-cyclohexylpicolinamide

[0171]

Production Example 92

3-Hydroxy-4,6-dimethoxy-4'-phenoxypicolinanilide

[0172]

Production Example 93

3-Hydroxy-4,5-dimethoxy-4'-phenoxypicolinanilide

[0173]

Production Example 94

3-Benzyloxy-4-methoxypicolinic acid:

3-Hydroxy-2-methyl-4-pyrone 25 g (0.198 mol) was dissolved in 70 ml of DMF. To the solution was added 8.7 g (0.218 mol) of sodium hydride (60% in mineral oil). The mixture was stirred under ice cooling for 30 min. Benzyl bromide 37.3 g (0.218 mol) was added dropwise to the reaction solution under ice cooling, and a reaction was allowed to proceed at room temperature overnight. The reaction solution was poured into ice water, followed by extraction with ethyl acetate. The organic layer was washed with water, and was dried over anhydrous sodium sulfate. The solvent was removed by distillation under the reduced pressure. The reddish brown oil thus obtained 64 g was applied to column chromatography on silica gel (Wako Gel C-200, n-hexane-ethyl acetate) to give 41.6 g (yield 97%) of 3-benzyloxy-2-methyl-4-pyrone.

^{1}H-NMR ($CDCl_3$): δ = 2.07 (s, 3H), 5.14 (s, 2H), 6.35 (1H, d), 7.28-7.39 (m, 5H), 7.58 (d, 1H)

[0174]

28% aqueous ammonia 100 ml and 30 ml of ethanol were added to 3-benzyloxy-2-methyl-4-pyrone 40.6 g (0.188 mol). The mixture was stirred at room temperature for 5 days. The reaction mixture was concentrated under the reduced pressure. The precipitate was filtered, and was then washed with a minor amount of ethyl acetate to give 32.2 g of 3-benzyloxy-2-methyl-4-pyridone as a light yellow crystal. The same compound was also obtained from the filtrate 5.6 g (yield 93%).

^{1}H-NMR ($CDCl_3$): δ = 2.13 (s, 3H), 5.02 (s, 2H), 6.32 (d, 1H), 7.22-7.30 (m, 5H), 7.37 (d, 1H), 13.13 (br, 1H)

[0175]

3-Benzyloxy-2-methyl-4-pyridone 21.5 g (0.10 mol) was suspended in methanol-acetonitrile (1 : 9 v/v) 400 ml. Diisopropylethylamine 18.1 g (0.14 mol) was added to the suspension. The mixture was then stirred. A 2.0 M solution 70 ml of tetramethylsilyldiazomethane in n-hexane was added dropwise to the mixture, and a reaction was allowed to proceed at room temperature overnight. The reaction solution was concentrated under the reduced pressure. The concentrate was applied to column chromatography on silica gel (Wako Gel C-200, n-hexane-ethyl acetate) to give 17.3 g (yield 76%) of 3-benzyloxy-4-methoxy-2-methylpyridine.

^{1}H-NMR ($CDCl_3$): δ = 2.34 (s, 3H), 3.84 (s, 3H), 4.91 (s, 2H), 6.66 (1H,d), 7.24-7.38 (m, 5H), 8.08 (d, 1H)

[0176]

3-Benzyloxy-4-methoxy-2-methylpyridine 23.0 g was dissolved in 200 ml of dichloromethane. m-Chloroperbenzoic acid 20.7 g was added to the solution under ice cooling, and a reaction was allowed to proceed at room temperature overnight. The reaction product was washed with an aqueous saturated sodium hydrogensulfite solution and an aqueous saturated sodium hydrogencarbonate solution, and the washed reaction product was dried over anhydrous sodium sulfate. The solvent was concentrated under the reduced pressure. Acetic anhydride 200 ml was added to 35.5 g of the concentrate as a light yellow oil, and a reaction was allowed to proceed at 100℃ for one hr. Ethanol 100 ml was then added thereto, and the mixture was further refluxed for one hr.

The reaction solution was concentrated under the reduced pressure. A 2 M solution 200 ml of sodium hydroxide in 50% methanol was added to the concentrate, and the mixture was stirred at 80℃ for one hr. The reaction solution was concentrated under the reduced pressure. The concentrate was extracted with chloroform. The extract was washed with saturated brine, and was then dried over anhydrous sodium sulfate, followed by concentration under the reduced pressure to give 19.6 g (yield 80%) of 3-benzyloxy-2-hydroxymethyl-4-methoxypyridine as a yellowish brown solid.

^{1}H-NMR ($CDCl_3$): δ = 3.89 (s, 3H), 4.56 (s, 2H), 4.97 (s, 2H), 6.77 (d, 1H), 7.24-7.36 (m, 5H), 8.15 (d, 1H)

[0177]

3-Benzyloxy-2-hydroxymethyl-4-methoxypyridine 7.1 g and 2.5 g of potassium hydroxide were suspended in 100 ml of water. While heating the suspension in a water bath, potassium permanganate 7.3 g was added thereto, and the mixture was stirred. The precipitate was filtered, and was washed with 100 ml of methanol. The filtrate and the washings were combined, followed by concentration under the reduced pressure. The concentrate was adjusted to pH 1 by the addition of concentrated hydrochloric acid. The precipitate was filtered, washed with water, and then dried to give 6.3 g (yield 83.9%) of 3-benzyloxy-4-methoxypiclinic acid as colorless powder.

[0178]

Production Example 95

3-Hydroxy-4-methoxypicolinic acid:

3-Benzyloxy-4-methoxypicolinic acid 5.3 g was suspended in 25 ml of ethanol. 10% palladium-carbon 0.5 g was added to the suspension. The mixture was then catalytically hydrogenated under atmospheric pressure for 30 min. The reaction solution was filtered under the reduced pressure. The filtrate was concentrated under the reduced pressure to give 2.8 g (yield 81.6%) of 3-hydroxy-4-methoxypicolinic acid as colorless powder.

[0179]

Production Example 96

3-Hydroxy-4-methoxypicolinic acid hydrochloride:

3-Benzyloxy-4-methoxypicolinic acid 8.3 g was dissolved in 100 ml of methanol. Concentrated hydrochloric acid 2 ml was added to the solution. The mixture was heated under reflux for 30 min. The reaction solution was concentrated under the reduced pressure. The residue was recrystallized from water-ethanol to give 3.6 g (yield 54.8%) of 3-hydroxy-4-methoxypicolic acid hydrochloride as colorless powder.

[0180]

Production Example 97

Methyl 3-benzyloxy-6-methoxy-picolinate:

3-Hydroxypicolinic acid 5.0 g was dissolved in 350 ml of toluene and 100 ml of methanol. A 2 M solution 25 ml of trimethylsilyldiazomethane in hexane was added dropwise to the solution, and a reaction was allowed to proceed at room temperature overnight. The reaction solution was concentrated under the reduced pressure. Methylene chloride 100 ml and water 100 ml were then added to the concentrate to conduct extraction. The aqueous layer was then extracted with methylene chloride. The organic layer was dried over magnesium sulfate, and was concentrated under the reduced pressure. The residue was purified by column chromatography on silica gel (ethyl acetate-hexane) to give 2.3 g (yield 41%) of methyl 3-hydroxy-picolinate.

^{1}H-NMR ($CDCl_3$): δ = 4.06 (s, 3H), 7.37 (dd, 1H), 7.43 (dd, 1H), 8.28 (dd, 1H)

[0181]

Methyl 3-hydroxypicolinate 2.0 g was dissolved in 100 ml of acetone. Potassium carbonate 3.4 g and 3.4 ml of benzyl bromide were added to the solution, and a reaction was allowed to proceed at room temperature overnight. The reaction solution was then refluxed for 4 hr. Water 50 ml was added thereto, and the mixture was neutralized with 1 N hydrochloric acid, followed by concentration under the reduced pressure. Methylene chloride and water were added to the residue. The organic layer was dried over magnesium sulfate, and was then dried under the reduced pressure. The dried organic layer was then purified by column chromatography (chloroform-methanol) to give 2.1 g (yield 62%) of methyl 3-benzyloxypicolinate.

^{1}H-NMR ($CDCl_3$): δ = 3.99 (s, 3H), 5.22 (s, 2H), 7.29 -7.48 (m, 7H), 8.29 (t, 1H)

[0182]

Methyl 3-benzyloxypicolinate 2.0 g was converted to N-oxide in the same manner as in Production Example 94, followed by acetylation to give methyl 6-acetoxy-3-benzyloxypicolinate which was then hydrolyzed with an alkali to give 0.77 g (yield 36%) of methyl 3-benzyloxy-6-hydroxy-picolinate.

^{1}H-NMR ($CDCl_3$): δ = 3.93 (s, 3H), 5.06 (s, 2H), 6.77 (d, 1H), 7.34 - 7.44 (m, 6H)

[0183]

Methyl 3-benzyloxy-6-hydroxy-picolinate 0.55 g was dissolved in 55 ml of acetone and 20 ml of methyl iodide. Potassium carbonate 1.4 g was added to the solution, and the mixture was refluxed for 3 hr. After cooling, the reaction solution was neutralized with 1 N hydrochloric acid, and was concentrated under the reduced pressure. Methylene chloride and water were then added to the concentrate to conduct extraction. The organic layer was dried over magnesium sulfate, and the dried organic layer was then concentrated under the reduced pressure. The residue was purified by column chromatography (chloroform-methanol) to give 0.28 g (yield 49%) of methyl 3-benzyloxy-6-methoxy-picolinate.

[0184]

Production Example 98

3-Benzyloxy-6-methoxy-picolinic acid:

Methyl 3-benzyloxy-6-methoxy-picolinate 20 mg was dissolved in 1 ml of methanol. A 1 N aqueous sodium hydroxide solution 0.33 ml was added to the solution, and a reaction was allowed to proceed at room temperature for 3 hr. The reaction solution was then adjusted to pH 3 by the addition of 1 N hydrochloric acid. The precipitate was collected by filtration to give 12 mg (yield 63%) of 3-benzyloxy-6-methoxy-picolinic acid.

[0185]

Production Example 99

Methyl 3-hydroxy-6-methoxy-picolinate:

10% palladium-carbon 48 mg was added to 480 mg of methyl

3-benzyloxy-6-methoxy-picolinate. After the replacement of the atmosphere by nitrogen, 25 ml of methanol was added thereto. Further, after the replacement of the atmosphere by hydrogen, the mixture was vigorously stirred to allow a reaction to proceed. One hr after the initiation of the reaction, the reaction mixture was filtered, followed by purification by chromatography on silica gel (chloroform-methanol) to give 240 mg (yield 76%) of methyl 3-hydroxy-6-methoxy-picolinate.

[0186]

Production Example 100

3-Hydroxy-6-methoxypicolinic acid:

Methyl 3-hydroxy-6-methoxypicolinate 80 mg was dissolved in 4 ml of methanol. A 1 N aqueous sodium hydroxide solution (2 ml) was added to the solution, and a reaction was allowed to proceed at room temperature for 3 hr. The reaction solution was adjusted to pH 3 by the addition of 1 N hydrochloric acid. The precipitate was collected by filtration to give 56 mg (yield 76%) of 3-hydroxy-6-methoxypicolinic acid.

[0187]

Production Example 101

Methyl 3-benzyloxy-4,6-dimethoxypicolinate:

3-Benzyloxy-4-methoxypicolinic acid (the compound of Production Example 94) 1 g was converted to a methyl ester in the same manner as in Production Example 97 to give 0.86 g (yield 81%) of methyl 3-benzyloxy-4-methoxypicolinate.

^{1}H-NMR ($CDCl_3$): δ = 3.82 (s, 3H), 3.83 (s, 3H), 5.02 (s, 2H), 6.86 (d, 1H), 7.19 - 7.41 (m, 5H), 8.22 (d, 1H)

[0188]

Methyl 3-benzyloxy-4-methoxypicolinate 0.80 g was oxidized with m-chloroperbenzoic acid in the same manner as in Production Example 94 to give 0.69 g (yield 81%) of methyl-N-oxide 3-benzyloxy-4-methoxypicolinate.

^{1}H-NMR ($CDCl_3$): δ = 3.83 (s, 3H), 3.86H (s, 3H), 5.04 (s, 2H), 6.74 (d, 1H), 7.19 - 7.41 (m, 5H), 7.91 (d, 1H)

[0189]

Methyl-N-oxide 3-benzyloxy-4-methoxypicolinate 672 mg was dissolved in

33.6 ml of acetic anhydride, and a reaction was allowed to proceed at 100°C overnight, followed by concentration under the reduced pressure. The concentrate was purified by chromatography on silica gel (ethyl acetate-hexane = 1 : 1) to give 173 mg (yield 22%) of methyl 6-acetoxy-3-benzyloxy-4-methoxypicolinate and 87 mg (yield 11%) of methyl 5-acetoxy-3-benzyloxy-4-methoxypicolinate.

Methyl 6-acetoxy-3-benzyloxy-4-methoxypicolinate

^{1}H-NMR ($CDCl_3$): δ = 2.25 (s, 3H), 3.82 (s, 3H), 3.88 (s, 3H), 5.02 (s, 2H), 6.71 (s, 1H), 7.19 - 7.43 (m, 5H)

Methyl 5-acetoxy-3-benzyloxy-4-methoxypicolinate

H-NMR ($CDCl_3$): δ = 2.37 (s, 3H), 3.92 (s, 3H), 4.00 (s, 3H), 5.10 (s, 2H), 7.19 - 7.43 (m, 5H), 8.19 (s, 1H)

[0190]

Methyl 6-acetoxy-3-benzyloxy-4-methoxypicolinate was hydrolized with an alkali in the same manner as in Production Example 97 to give 96 mg (yield 85%) of methyl 3-benzyloxy-6-hydroxy-4-methoxypicolinate.

^{1}H-NMR ($CDCl_3$): δ = 3.80 (s, 3H), 3.81 (s, 3H), 4.87 (s, 2H), 6.04 (s, 1H), 7.19 - 7.37 (m, 5H), 9.39 (br, 1H)

[0191]

Methyl 3-benzyloxy-6-hydroxy-4-methoxypicolinate 90 mg was methylated in the same manner as in Production Example 97 to give 33 mg (yield 35%) of methyl 3-bynzyloxy-4,6-dimethoxypicolinate.

[0192]

Production Example 102

3-Benzyloxy-4,6-dimethoxypicolinic acid:

Methyl 3-benzyloxy-4,6-dimethoxypicolinate 33 mg was dissolved in 2 ml of methanol. A 1 N aqueous sodium hydroxide solution 0.54 ml was added to the solution, and a reaction was allowed to proceed at room temperature for 4 hr. The reaction solution was neutralized with 1 N hydrochloric acid, and was then concentrated under the reduced pressure to give 3-benzyloxy-4,6-dimethoxypicolinic acid.

[0193]

Production Example 103

Methyl 3-benzyloxy-4,5-dimethoxypicolinate:

Methyl 5-acetoxy-3-benzyloxy-4-methoxypicolinate 87 mg was hydrolyzed with an alkali in the same manner as in Production Example 101 to give 71 mg (yield 93%) of methyl 3-benzyloxy-5-hydroxy-4-methoxypicolinate.

^{1}H-NMR ($CDCl_3$): δ = 3.84 (s, 3H), 3.98 (s, 3H), 5.01 (s, 2H), 7.19 - 7.42 (m, 5H), 8.12 (s, 1H)

[0194]

The procedure of Production Example 101 was repeated, except that 71 mg of methyl 3-benzyloxy-5-hydroxy-4-methoxypicolinate was used. Thus, 21 mg (yield 28%) of methyl 3-benzyloxy-4,5-dimethoxypicolinate.

[0195]

Production Example 104

3-Benzyloxy-4,5-dimethoxypicolinic acid:

Methyl 3-benzyloxy-4,5-dimethoxypicolinate 20 mg was dissolved in 1 ml of methanol. A 1 N aqueous sodium hydroxide solution 0.33 ml was added to the solution, and a reaction was allowed to proceed at room temperature for 3 hr. The reaction solution was neutralized with 1 N hydrochloric acid, and was then concentrated under the reduced pressure to give 3-benzyloxy-4,5-dimethoxypicolinic acid.

[0196]

NMR spectral data on the compounds of the present invention were as shown in Tables 1 to 5 below. In the tables, c represents $CDCl_3$, d DMSO-d_6, m methanol-d_4, and w D_2O.

[0197]

[Table 1]

Comp. No.	^{1}H-NMR δ (ppm) Table 1	Solvent for measurement
1	6.94-7.06 (m, 5H), 7.25-7.35 (m, 4H), 7.59-7.63 (m, 2H), 8.06 (dd, 1H),	c
	9.82 (s, 1H), 11.86 (s, 1H)	
2	3.97 (s, 2H), 7.17-7.22 (m, 5H), 7.26-7.39 (m, 4H), 7.61 (m, 2H), 8.10 (dd, 1H),	c
	9.85 (s, 1H), 11.94 (s, 1H)	
3	0.72-0.82 (m, 6H), 1.16-1.23 (m, 6H), 1.50-1.56 (m, 4H), 2.77-2.81 (m, 2H),	c
	6.78-7.37 (m, 7H), 7.55-7.61 (m, 2H), 8.09 (dd, 1H), 9.81 (s, 1H), 11.97 (s, 1H)	
4	1.32 (s, 9H), 6.95 (d, 2H), 7.05 (d, 2H), 7.35 (d, 2H), 7.36 (dd, 1H), 7.40 (dd, 1H),	c
	7.66 (d, 2H), 8.13 (dd, 1H), 9.88 (br, 1H), 11.95 (s, 1H)	
5	1.33 (s, 9H), 1.42 (s, 9H), 6.76 (d, 1H), 7.02 (d, 2H), 7.14 (dd, 1H), 7.36 (dd, 1H),	c
	7.40 (dd, 1H), 7.41 (d, 1H), 7.64 (d, 2H), 8.13 (dd, 1H), 9.86 (br, 1H), 11.98 (s, 1H)	
6	7.09 (d, 2H), 7.17 (d, 1H), 7.37 (dd, 1H), 7.30-7.46 (m, 3H), 7.42 (dd, 1H),	c
	7.73 (d, 2H), 8.13 (dd, 1H), 9.94 (br, 1H), 11.88 (s, 1H)	
7	1.23-1.49 (m, 5H), 1.64 (m, 1H), 1.79 (m, 2H), 2.02 (m, 2H), 3.92 (m, 1H),	c
	7.29 (dd, 1H), 7.33 (dd, 1H), 7.93 (br, 1H), 8.04 (dd, 1H), 12.33 (s, 1H)	
8	4.01 (s, 3H), 5.05 (s, 2H), 6.93-7.12 (m, 6H), 7.27-7.48 (m, 7H),	d
	7.74-7.78 (m, 2H), 8.31 (d, 1H), 10.46 (s, 1H)	
9	3.96 (s, 3H), 6.89-7.10 (m, 6H), 7.24-7.34 (m, 2H), 7.64-7.67 (m, 2H),	c
	8.01 (d, 1H), 9.90 (s, 1H), 12.17 (s, 1H)	
10	1.33 (s, 9H), 3.98 (s, 3H), 6.91 (d, 1H), 6.95 (d, 2H), 7.02 (d, 2H), 7.35 (d, 2H),	c
	7.65 (d, 2H), 8.03 (d, 1H), 9.91 (br, 1H), 12.20 (s, 1H)	
11	3.90 (s, 3H), 6.75-7.08 (m, 5H), 7.25-7.45 (m, 5H), 7.94 (d, 1H), 9.87 (s, 1H),	c
	12.01 (s, 1H)	
12	3.89 (s, 3H), 6.80-7.12 (m, 7H), 7.29-7.33 (m, 2H), 7.91 (d, 1H), 8.48 (d, 1H),	c
	10.51 (s, 1H), 12.09 (s, 1H)	
13	3.95 (s, 3H), 3.97 (s, 2H), 6.89 (d, 1H), 7.16-7.29 (m, 7H), 7.60 (d, 2H),	c
	8.00 (d, 1H), 9.88 (s, 1H), 12.20 (s, 1H)	
14	3.96 (s, 3H), 6.94 (d, 1H), 7.18-7.30 (m, 5H), 7.40 (d, 2H), 7.66 (d, 2H),	c
	8.01 (d, 1H), 9.97 (s, 1H), 12.05 (s, 1H)	
15	3.79 (s, 3H), 3.95 (s, 3H), 6.86-6.98 (m, 7H), 7.61 (d, 2H), 8.00 (d, 1H),	c
	9.87 (s, 1H), 12.19 (s, 1H)	

[0198]

[Table 2]

Comp. No.	^{1}H-NMR δ (ppm) Table 2	Solvent for measurement
16	3.97 (s, 3H), 6.92 (d, 1H), 7.04-7.07 (m, 3H), 7.59 (d, 2H), 7.92 (m, 1H),	c
	8.03 (d, 1H), 8.06 (d, 1H), 10.08 (s, 1H), 11.85 (s, 1H)	
17	3.96 (s, 3H), 6.90 (d, 1H), 7.04-7.10 (m, 4H), 7.29-7.35 (m, 1H),	c
	7.40-7.43 (m, 2H), 7.50-7.56 (m, 4H), 7.67-7.69 (m, 2H), 8.01 (d, 1H), 9.92 (s, 1H),	
	12.17 (s, 1H)	
18	2.34 (s, 3H), 3.98 (s, 3H), 6.91 (d, 1H), 6.92 (d, 2H), 7.02 (d, 2H), 7.12 (d, 2H),	c
	7.65 (d, 2H), 8.03 (d, 1H), 9.90 (br, 1H), 12.20 (s, 1H)	
19	2.35 (s, 3H), 3.98 (s, 3H), 6.92-6.98 (m, 4H), 7.17 (d, 2H), 7.81 (dd, 1H),	c
	8.01 (d, 1H), 8.04 (d, 1H), 10.00 (br, 1H), 11.96 (s, 1H)	
20	3.92 (s, 3H), 3.97 (s, 3H), 6.64 (dd, 1H), 6.69 (d, 1H), 6.91 (d, 1H), 7.02 (dd, 2H),	c
	7.11 (m, 1H), 7.34 (dd, 2H), 8.07 (d, 1H), 8.38 (d, 1H), 10.38 (br, 1H), 12.30 (s, 1H)	
21	3.98 (s, 3H), 6.91-7.29 (m, 5H), 7.33 (m, 2H), 7.53 (m, 1H), 7.97 (d, 1H), 8.03 (d, 1H),	c
	9.97 (br, 1H), 11.99 (s, 1H)	
22	3.98 (s, 3H), 6.93 (d, 1H), 6.99 (d, 1H), 7.04 (dd, 2H), 7.16 (t, 1H), 7.37 (dd, 2H),	c
	7.84 (dd, 1H), 8.03 (d, 1H), 8.04 (d, 1H), 10.02 (br, 1H), 11.94 (s,1H)	
23	2.27 (s, 3H), 3.98 (s, 3H), 6.91 (m,1H), 6.91 (dd, 2H), 6.95 (d, 1H), 7.05 (t, 1H),	c
	7.31 (dd, 2H), 7.49 (dd, 1H), 7.64 (d, 1H), 8.03 (d, 1H), 9.91 (br, 1H), 12.21 (s, 1H)	
24	2.34 (s, 3H), 3.91 (s, 3H), 3.97 (s, 3H), 6.60 (dd, 1H), 6.66 (d, 1H), 6.90 (d, 1H),	c
	6.92 (d, 2H), 7.14 (d, 2H), 8.06 (d, 1H), 8.34 (d, 1H), 10.36 (br, 1H), 12.31 (s, 1H)	
25	1.32 (s, 9H), 1.42 (s, 9H), 3.98 (s, 3H), 6.75 (d, 1H), 6.91 (d, 1H), 7.01 (d, 2H),	c
	7.14 (dd, 1H), 7.41 (d, 1H), 7.64 (d, 2H), 8.03 (d, 1H), 9.89 (br, 1H), 12.23 (s, 1H)	
26	3.98 (s, 3H), 6.93 (d, 1H), 7.08 (d, 2H), 7.16 (d, 1H), 7.26 (s, 1H), 7.34 (d, 1H),	c
	7.43 (dd, 1H), 7.73 (d, 2H), 8.04 (d, 1H), 9.97 (br, 1H), 12.13 (s, 1H)	
27	1.30 (s, 18H), 3.98 (s, 3H), 6.88 (d, 2H), 6.91 (d, 1H), 7.04 (d, 2H), 7.18 (t, 1H),	c
	7.66 (d, 2H), 8.03 (d, 1H), 9.92 (br, 1H), 12.22 (s, 1H)	
28	3.98 (s, 3H), 6.89 (d, 2H), 6.93 (d, 1H), 7.04 (d, 1H), 7.28 (d, 2H), 7.55 (dd, 1H),	c
	7.97 (d, 1H), 8.03 (d, 1H), 9.98 (br, 1H), 11.95 (s, 1H)	
29	2.28 (s, 3H), 3.78 (s, 3H), 3.95 (s, 3H), 6.80-6.90 (m, 6H), 7.41 (dd, 1H),	c
	7.59 (d, 1H), 8.00 (d, 1H), 9.85 (s, 1H), 12.21 (s, 1H)	
30	1.77 (d, 3H), 3.92 (s, 3H), 6.05 (t, 1H), 6.83 (d, 1H), 7.44-7.59 (m, 4H), 7.80 (d, 1H),	c
	7.86 (d, 1H), 7.89 (d, 1H), 8.14 (d, 1H), 8.33 (br, 1H), 12.41 (s, 1H)	

[0199]

[Table 3]

Comp. No.	^{1}H-NMR δ (ppm) Table 3	Solvent for measurement
31	3.79 (s, 3H), 3.96 (s, 3H), 6.85-6.98 (m, 6H), 7.45 (dd, 1H), 7.91 (d, 1H),	c
	8.00 (d, 1H), 9.91 (br, 1H), 11.99 (s, 1H)	
32	2.33 (s, 3H), 3.98 (s, 3H), 6.88 (d, 2H), 6.92 (d, 1H), 6.98 (d, 1H), 7.14 (d, 2H),	c
	7.50 (dd, 1H), 7.95 (d, 1H), 8.03 (d, 1H), 9.95 (br, 1H), 12.01 (s, 1H)	
33	2.28 (s, 3H), 2.32 (s, 3H), 3.98 (s, 3H), 6.82 (d, 2H), 6.91 (d, 1H), 6.92 (d, 1H),	c
	7.11 (d, 2H), 7.46 (dd, 1H), 7.62 (d, 1H), 8.03 (d, 1H), 9.89 (br, 1H), 12.22 (s, 1H)	
34	3.98 (s, 3H), 6.92 (d, 1H), 7.01 (d, 2H), 7.06 (d, 2H), 7.09 (d, 2H), 7.71 (d, 2H),	c
	8.04 (d, 1H), 9.95 (br, 1H), 12.14 (s, 1H)	
35	3.98 (s, 3H), 6.86 (m, 1H), 6.93 (d, 1H), 6.90-6.96 (m, 2H), 7.09 (d, 2H),	c
	7.33 (dd, 1H), 7.73 (d, 2H), 8.04 (d, 1H), 9.97 (br, 1H), 12.14 (s, 1H)	
36	2.36 (s, 3H), 3.98 (s, 3H), 6.93 (d, 1H), 6.94 (d, 2H), 7.18 (d, 2H), 7.19 (dd, 1H),	c
	7.30 (d, 1H), 8.07 (d, 1H), 8.23 (d, 1H), 10.37 (br, 1H), 11.85 (s, 1H)	
37	2.31 (s, 3H), 2.34 (s, 3H), 3.96 (s, 3H), 6.57 (d, 1H), 6.70 (dd, 1H), 6.87 (d, 2H),	c
	6.88 (d, 1H), 7.01 (d, 2H), 7.10 (d, 2H), 7.16 (d, 2H), 7.99 (d, 1H), 8.42 (d, 1H),	
	10.47 (br, 1H), 12.20 (s, 1H)	
38	1.36 (s, 18H), 3.98 (s, 3H), 6.91 (d, 1H), 7.25 (d, 1H), 7.56 (d, 2H), 8.04 (d, 1H),	c
	9.91 (br, 1H), 12.33 (s, 1H)	
39	3.95 (s, 3H), 5.06 (s, 2H), 6.89 (d, 1H), 6.97-7.00 (m, 2H), 7.29-7.43 (m, 5H),	c
	7.60 (d, 2H), 8.00 (d, 1H), 9.82 (s, 1H), 12.25 (s, 1H)	
40	3.95 (s, 3H), 5.09 (s, 2H), 6.79 (d, 1H), 6.89 (d, 1H), 7.17-7.45 (m, 7H),	c
	8.01 (d, 1H), 9.93 (br, 1H), 12.15 (s, 1H)	
41	3.97 (s, 3H), 6.92 (d, 1H), 7.36-7.40 (m, 2H), 7.49 (dd, 1H), 7.71 (d, 1H), 7.91 (m, 1H),	c
	7.99-8.04 (m, 2H), 8.60 (m, 1H), 8.87 (d, 1H), 10.06 (s, 1H), 12.08 (s, 1H)	
42	1.36-1.70 (m, 20H), 1.73 (m, 2H), 3.92 (s, 3H), 4.18 (m, 1H), 6.83 (d, 1H),	c
	7.85 (br, 1H), 7.92 (dd, 1H), 12.59 (s, 1H)	
43	1.45-1.72 (m, 12H), 1.91 (m, 2H), 3.92 (s, 3H), 4.12 (m, 1H), 6.83 (d, 1H),	c
	7.92 (dd, 1H), 7.97 (br, 1H), 12.60 (s, 1H)	
44	3.90 (s, 3H), 5.64 (br, 1H), 6.86 (m, 2H), 6.99 (d, 2H), 7.04 (d, 2H), 7.19 (m, 1H),	c
	7.54 (d, 2H), 7.96 (dd,1H), 9.79 (br, 1H), 12.23 (s, 1H)	
45	1.71 (m, 6H), 2.12 (m, 9H), 3.91 (s, 3H), 6.82 (d, 1H), 7.87 (br, 1H), 7.90 (dd, 1H),	c
	12.69 (s, 1H)	

[0200]

[Table 4]

Comp. No.	^{1}H-NMR δ (ppm) Table 4	Solvent for measurement
46	3.14 (m, 4H), 3.85 (m, 4H), 3.95 (s, 3H), 6.88 (d, 1H), 6.92 (d, 2H), 7.59 (d, 2H),	c
	8.00 (d, 1H), 9.80 (br, 1H), 12.29 (s, 1H)	
47	1.53 (m, 6H), 1.67 (m, 6H), 1.98 (m, 3H), 3.11 (d, 2H), 3.93 (s, 3H), 6.84 (d, 1H),	c
	7.95 (d, 1H), 8.13 (br, 1H), 12.55 (s, 1H)	
48	2.25 (s, 3H), 3.98 (s, 3H), 6.93 (d, 1H), 6.98 (d, 1H), 7.05 (d, 1H), 7.16 (m, 1H),	c
	7.28 (d, 1H), 7.40 (dd, 1H), 7.53 (dd, 1H), 7.68 (d, 1H), 8.04 (d, 1H), 9.95 (br, 1H),	
	12.15 (s, 1H)	
49	1.23-1.28 (m, 1H), 1.36-1.44 (m, 4H), 1.73-1.78 (m, 1H), 1.81-1.91 (m, 5H),	c
	3.97 (s, 3H), 6.91 (d, 1H), 7.24 (d, 2H), 7.61 (d, 2H), 8.03 (d, 1H), 9.89 (br, 1H),	
	12.28 (s, 1H)	
50	3.75 (m, 8H), 3.90 (m, 4H), 3.95 (s, 3H), 4.15 (m, 4H), 6.87 (m, 2H), 7.09 (dd, 1H),	c
	7.43 (d, 1H), 7.99 (d, 1H), 9.83 (br, 1H), 12.20 (s, 1H)	
51	3.95 (s, 3H), 4.25 (m, 4H), 6.85 (d, 1H), 6.88 (d, 1H), 7.08 (dd, 1H), 7.32 (m, 1H),	c
	7.99 (dd, 1H), 9.77 (br, 1H), 12.23 (s, 1H)	
52	1.63 (m, 2H), 1.98 (m, 2H), 2.17 (m, 2H), 2.84 (m, 2H), 3.50 (s, 2H), 3.92 (s, 3H),	c
	3.92 (m, 1H), 6.84 (d, 1H), 7.24 (m, 1H), 7.30 (d, 4H), 7.93 (dd, 1H),	
	7.93 (br, 1H), 12.47 (s, 1H)	
53	1.53 (m, 2H), 1.63 (m, 2H), 1.97 (m, 4H), 2.23 (t, 2H), 3.49 (m, 2H), 3.92 (s, 3H),	c
	5.51 (s, 1H), 6.83 (d, 1H), 7.92 (dd, 1H), 8.01 (br, 1H), 12.52 (s, 1H)	
54	3.99 (s, 3H), 6.94 (d, 1H), 7.04 (d, 2H), 7.14 (d, 2H), 7.79 (d, 2H), 8.05 (d, 1H),	c
	8.22 (d, 2H), 10.02 (br, 1H), 12.06 (s, 1H)	
55	2.26 (s, 6H), 3.98 (s, 3H), 6.78 (s, 2H), 6.93 (d, 1H), 7.04 (d, 2H), 7.11 (t, 1H),	c
	7.35 (t, 2H), 8.05 (d, 1H), 9.34 (br, 1H), 12.27 (s, 1H)	
56	1.32-1.40 (m, 2H), 2.21-2.25 (m, 1H), 3.06-3.11 (m, 1H), 3.94 (s, 3H), 6.87 (d, 1H),	c
	7.18-7.22 (m, 3H), 7.30 (t, 2H), 7.95 (d, 1H), 8.19 (br, 1H), 12.36 (s, 1H)	
57	1.45-1.70 (m, 10H), 2.00 (m, 2H), 3.92 (s, 3H), 4.08 (m, 1H), 6.83 (d, 1H),	c
	7.93 (dd, 1H), 7.96 (br, 1H), 12.60 (s, 1H)	
58	1.22 (d, 6H), 3.59 (m, 1H), 3.97 (s, 3H), 6.58 (d, 2H), 6.89 (d, 2H), 6.91 (d, 1H),	c
	6.96 (d, 2H), 7.60 (d, 2H), 8.02 (d, 1H), 9.86 (br, 1H), 12.24 (s, 1H)	
59	1.17-1.51 (m, 5H), 1.64 (m, 1H), 1.77 (m, 2H), 1.98 (m, 2H), 3.89 (m, 1H),	c
	3.92 (s, 3H),6.83 (d, 1H), 7.92 (br, 1H), 7.93 (dd, 1H), 12.60 (s, 1H)	
60	3.98 (s, 3H), 6.92 (d, 1H), 7.19 (t, 1H), 7.40 (t, 2H), 7.72 (d, 2H), 8.04 (d, 1H),	c
	9.96 (br, 1H), 12.20 (s, 1H)	

[0201]

[Table 5]

Comp. No.	^{1}H-NMR δ (ppm) Table 5	Solvent for measurement
61	3.98 (s, 3H), 6.93 (d, 1H), 7.36 (d, 2H), 7.68 (d, 2H), 8.03 (d, 1H), 9.97 (br, 1H),	c
	12.04 (s, 1H)	
62	3.97 (s, 3H), 6.69 (d, 2H), 6.88 (d, 2H), 6.91 (d, 1H), 6.97 (d, 2H), 7.61 (d, 2H),	c
	8.02 (d, 1H), 9.87 (br, 1H), 12.23 (s, 1H)	
63	0.89 (m, 2H), 1.13 (m, 2H), 1.30 (m, 1H), 1.45 (m, 4H), 1.64 (m, 4H), 3.39 (m, 2H),	c
	3.88 (s, 3H), 6.79 (d, 1H), 7.88 (d, 1H), 7.92 (br, 1H), 12.49 (s, 1H)	
64	3.92 (s, 3H), 6.88 (d, 1H), 7.43 (t, 2H), 7.53 (m, 1H), 7.73 (m, 2H), 7.78 (m, 2H),	c
	7.83 (m, 2H), 7.99 (d, 1H), 10.13 (br, 1H), 11.87 (s, 1H)	
65	1.96-2.05 (m, 1H), 2.64-2.72 (m, 1H), 2.91-2.99 (m, 1H), 3.04-3.12 (m, 1H),	c
	3.96 (s, 3H), 5.60-5.66 (m, 1H), 6.86 (d, 1H), 7.22-7.28 (m, 3H), 7.36 (d, 1H),	
	7.92 (d, 1H), 8.24 (br, 1H), 12.49 (s, 1H)	
66	1.88-2.00 (m, 3H), 2.13-2.20 (m, 1H), 2.79-2.91 (m, 2H), 3.95 (s, 3H),	c
	5.30-5.36 (m, 1H), 6.85 (d, 1H), 7.13-7.22 (m, 3H), 7.31 (d, 1H), 7.91 (d, 1H),	
	8.29 (br, 1H), 12.53 (s, 1H)	
67	3.95 (s, 3H), 4.64 (d, 2H), 6.86 (d, 1H), 7.28-7.38 (m, 5H), 7.94 (d, 1H),	c
	8.36 (br, 1H), 12.38 (s, 1H)	
68	2.95 (t, 2H), 3.70 (t, 2H), 3.94 (s, 3H), 6.85 (d, 1H), 7.23-7.26 (m, 3H),	c
	7.31-7.34 (m, 2H), 7.92 (d, 1H), 8.12 (br, 1H), 12.44 (s, 1H)	
69	1.63 (d, 3H), 3.94 (s, 3H), 5.25 (q, 1H), 6.86 (d, 1H), 7.28 (m, 1H), 7.34-7.41 (m, 4H),	c
	7.95 (d, 1H), 8.31 (br, 1H), 12.38 (s, 1H)	
70	1.83 (s, 6H), 3.93 (s, 3H), 6.86 (d, 1H), 7.26 (t, 1H), 7.35 (t, 2H), 7.45 (d, 2H),	c
	7.96 (d, 1H), 8.48 (br, 1H), 12.35 (s, 1H)	
71	3.95 (s, 3H), 4.60 (d, 2H), 6.87 (d, 1H), 6.97-7.02 (m, 4H), 7.10 (t, 1H),	c
	7.31-7.35 (m, 4H), 7.95 (d, 1H), 8.34 (br, 1H), 12.37 (s, 1H)	
72	3.11 (t, 2H), 3.97 (s, 3H), 4.19 (t, 2H), 6.90 (d, 1H), 6.92 (d, 2H), 7.23-7.35 (m, 5H),	c
	7.60 (d, 2H), 8.02 (d, 1H), 9.83 (br, 1H), 12.27 (s, 1H)	
73	1.16 (d, 6H), 2.84 (m, 1H), 3.16 (br, 4H), 3.69 (br, 2H), 3.80 (br, 2H), 3.97 (s, 3H),	c
	6.90 (d, 1H), 6.96 (d, 2H), 7.62 (d, 2H), 8.02 (d, 1H), 9.84 (br, 1H), 12.28 (s, 1H)	
74	1.61 (m, 4H), 1.72 (m, 4H), 3.06 (t, 4H), 3.87 (s, 3H), 6.79 (d, 1H), 7.86 (d, 1H),	c
	8.94 (br, 1H), 12.30 (s, 1H)	
75	0.97-1.07 (m, 2H), 1.13-1.30 (m, 3H), 1.55-1.64 (m, 1H), 1.66-1.69 (m, 1H),	c
	1.73-1.81 (m, 4H), 3.28 (t, 2H), 3.94 (s, 3H), 6.86 (d, 1H), 7.95 (d, 1H), 8.11 (br, 1H),	
	12.55 (s, 1H)	

[0202]

[Table 6]

Comp. No.	^{1}H-NMR δ (ppm) Table 6	Solvent for measurement
76	0.97 (d, 3H), 1.13-1.19 (m, 1H), 1.23-1.33 (m, 2H), 1.35-1.44 (m, 2H),	c
	1.69-1.73 (m, 1H), 1.78-1.84 (m, 2H), 2.01-2.05 (m, 1H), 3.58-3.62 (m, 1H),	
	3.94 (s, 3H), 6.86 (d, 1H), 7.86 (br, 1H), 7.95 (d, 1H), 12.64 (s, 1H)	
77	0.94 (d, 3H), 1.34-1.43 (m, 2H), 1.53-1.70 (m, 5H), 1.77-1.83 (m, 1H),	c
	1.90-1.96 (m, 1H), 3.94 (s, 3H), 4.17-4.22 (m, 1H), 6.86 (d, 1H), 7.96 (d, 1H),	
	8.21 (br, 1H), 12.61 (s, 1H)	
78	0.92 (d, 3H), 0.97 (d, 3H), 1.05-1.16 (m, 2H), 1.25-1.40 (m, 6H), 1.58 (m, 1H),	c
	1.63-1.83 (m, 8H), 2.02-2.08 (m, 1H), 3.80-3.88 (m, 1H), 3.94 (s, 3H), 3.95 (s, 3H),	
	4.12-4.17 (m, 1H), 6.85 (d, 1H), 6.86 (d, 1H), 7.87 (br, 1H), 7.94 (d, 1H),	
	7.97 (d, 1H), 8.20 (br, 1H), 12.60 (s, 1H), 12.61 (br, 1H)	
79	1.59-1.62 (m, 2H), 1.64-1.72 (m, 2H), 1.76-1.79 (m, 2H), 2.04-2.10 (m, 2H),	c
	3.94 (s, 3H), 4.33-4.40 (m, 1H), 6.85 (d, 1H), 7.94 (d, 1H), 7.94 (br, 1H), 12.59 (s, 1H)	
80	0.70 (m, 2H), 0.89 (m, 2H), 2.90 (m, 1H), 3.94 (s, 3H), 6.86 (d, 1H), 7.93 (d, 1H),	c
	8.03 (br, 1H), 12.42 (s, 1H)	
81	1.77-1.80 (m, 2H), 2.02-2.12 (m, 2H), 2.39-2.46 (m, 2H), 3.94 (s, 3H), 4.53 (m, 1H),	c
	6.86 (d, 1H), 7.95 (d, 1H), 8.14 (br, 1H), 12.47 (s, 1H)	
82	0.97 (t, 3H), 1.26 (d, 3H), 1.61 (q, 2H), 3.94 (s, 3H), 4.00-4.12 (m, 1H),	c
	6.86 (d, 1H), 7.86 (br, 1H), 7.95 (d, 1H), 12.61 (s, 1H)	
83	0.89 (t, 3H), 1.30-1.34 (m, 4H), 1.36-1.42 (m, 2H), 1.59-1.67 (m, 2H), 3.43 (q, 2H),	c
	3.94 (s, 3H), 6.86 (d, 1H), 7.95 (d, 1H), 8.04 (br, 1H), 12.55 (s, 1H)	
84	1.39-1.52 (m, 4H), 2.04-2.12 (m, 4H), 3.68 (m, 1H), 3.91 (m, 1H), 3.94 (s, 3H),	c
	6.86 (d, 1H), 7.89 (br, 1H), 7.94 (d, 1H), 12.49 (s, 1H)	
85	1.28-1.46 (m, 4H), 1.79 (m, 2H), 2.11 (m, 2H), 3.51 (m, 1H), 3.80 (m, 1H),	c
	3.95 (s, 3H), 6.87 (d, 1H), 7.96 (d, 1H), 8.05 (br, 1H), 12.26 (s, 1H)	
86	0.88 (t, 3H), 1.26-1.42 (m, 10H), 1.64 (m, 2H), 3.43 (m, 2H), 3.94 (s, 3H),	c
	6.86 (d, 1H), 7.95 (d, 1H), 8.03 (br, 1H), 12.55 (s, 1H)	
87	0.88 (t, 3H), 1.25-1.44 (m, 8H), 3.43 (m, 2H), 3.94 (m, 2H), 3.94 (s, 3H),	c
	6.86 (d, 1H), 7.95 (d, 1H), 8.03 (br, 1H), 12.54 (s, 1H)	
88	0.98 (s, 9H), 1.56 (t, 2H), 3.43-3.48 (m, 2H), 3.94 (s, 3H), 6.85 (d, 1H), 7.94 (d, 1H),	c
	7.98 (br, 1H), 12.53 (s, 1H)	
89	3.52 (s, 3H), 4.95 (s, 2H), 6.53 (d, 1H), 6.98-7.01 (m, 4H), 7.08-7.12 (m, 1H),	c
	7.26-7.35 (m, 8H), 7.47-7.50 (m, 2H), 8.31 (br, 1H)	
90	3.57 (s, 3H), 6.60 (d, 1H), 6.95-7.02 (m, 4H), 7.08-7.12 (m, 1H), 7.31-7.40 (m, 3H),	m
	7.65-7.69 (m, 2H)	

[0203]

[Table 7]

Comp. No.	^{1}H-NMR δ (ppm) Table 7	Solvent for measurement
91	1.12-1.48 (m, 5H), 1.68-2.17 (m, 5H), 3.55 (s, 3H), 3.94 (m, 1H), 6.57 (d, 1H),	c
	7.16 (d, 1H)	
94	4.05 (s, 3H), 5.15 (s, 2H), 7.28-7.37 (m, 4H), 7.47-7.50 (m, 2H), 8.25 (d, 1H)	m
95	4.03 (s, 3H), 7.39 (d, 1H), 8.04 (d, 1H)	d
96	4.14 (s, 3H), 7.46 (d, 1H), 8.08 (d, 1H)	m
97	3.44 (s, 3H), 3.90 (s, 3H), 4.93 (s, 2H), 6.60 (d, 1H), 7.25 (d, 1H),	c
	7.30～7.44 (m, 5H)	
98	3.51 (s, 3H), 5.04 (s, 2H), 6.52 (d, 1H), 7.40～7.45 (m, 5H), 7.61 (d, 1H)	w
100	3.37 (s, 3H), 6.41 (d, 1H), 7.21 (d, 1H)	w
101	3.80 (s, 3H), 3.81 (s, 3H), 3.85 (s, 3H), 4.92 (s, 2H), 6.27 (s, 1H),	c
	7.19～7.39 (m, 5H)	
103	3.91 (s, 3H), 4.00 (s, 3H), 4.01 (s, 3H), 5.11 (s, 2H), 7.19～7.42 (m, 5H),	c
	8.14 (s, 1H)	

[0204]

Preparation examples are given below. In these examples, the term "part" or "parts" means a part by weight or parts by weight.

[0205]

Preparation Example 1 Emulsifiable concentrate

Each compound 20 parts of the present invention was dissolved in 50 parts of xylene and 20 parts of DMF. Polyoxyethylene alkylaryl ether 10 parts was added to the solution, followed by mixing while stirring. Thus, 20% emulsifiable concentrates were prepared.

[0206]

Preparation Example 2 Wettable powder

Each compound 25 parts of the present invention was added to a mixture of 7 parts of polyoxyethylene alkylaryl ether, 3 parts of calcium ligninsulfonate, 30 parts of

clay, and 35 parts of diatomaceous earth, followed by homogeneous mixing while stirring in a juice mixer. Thus, 25% wettable powders were prepared.

[0207]

Preparation Example 3 Granules

Calcium ligninsulfonate 2 parts, 40 parts of bentonite, and 53 parts of talc were added to and thoroughly mixed with each compound 5 parts of the present invention while stirring. A suitable amount of water was then added to these mixtures, and the mixtures were stirred and thoroughly kneaded. The kneaded products were then granulated by means of a granulator, followed by forced draft drying to prepare 5% granules.

[0208]

Preparation Example 4 Dust

Each compound 2 parts of the present invention produced above was dissolved in a suitable amount of acetone. Talc 37 parts, 1 part of calcium stearate, and 60 parts of clay were added to the solutions, followed by mixing while stirring in a juice mixer. Acetone was removed by evaporation to prepare 2% dusts.

[0209]

Next, the fact that the compound of the present invention have the excellent control activity against plant pathogenic fungi will be described according to the following test examples.

[0210]

Test Example 1 Preventive effect against rice blast

The 20% emulsifiable concentrate prepared in Preparation Example 1 was diluted with water to prepare a test solution having a concentration of 100 ppm. The test solution was applied to stems and leaves of fourth-leaf stage rice seedlings (cultivar: Jikkoku) raised in an environment control room. The rice seedlings, to which the test solution had been applied, were air dried. Thereafter, the rice seedlings were inoculated by spraying with a conidial suspension of rice blast fungi (Pyricularia oryze). These rice seedlings were then allowed to stand within an inoculation box kept at a humidity of 100% for 40 hr after the inoculation to render the condition suitable for

infection, and were then transferred to an environment controlled greenhouse to induce the disease. Six days after the inoculation, the number of lesions per leaf was counted and compared with the number of lesions per leaf in the nontreated plot to calculate the protective value. The results were evaluated according to the following criteria.

A: Protective value 100% to 80%

B: Protective value 79% to 50%

C: Protective value 49% to 0%

[0211]

For the compounds produced in Production Examples 1, 4, 6, 9, 10, 11, 12, 13, 15, 16, 17, 18, 19, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 42, 43, 45, 47, 48, 49, 51, 53, 54, 55, 56, 57, 58, 59, 60, 61, 63, 68, 69, 70, 71, 72, 73, 75, 76, 77, 78, 79, 81, 82, 83, 86, and 88, the protective value was evaluated as A. These compounds had no phytotoxicity.

[0212]

Test Example 2 Preventive effect against wheat leaf rust

The 20% emulsifiable concentrate prepared in Preparation Example 1 was diluted with water to prepare a test solution having a concentration of 200 ppm. The test solution was applied to stems and leaves of fourth-leaf stage wheat seedlings (cultivar: Norin No. 61) raised in an environment controlled greenhouse. The wheat seedlings, to which the test solution had been applied, were air dried. Thereafter, the wheat seedlings were inoculated by spraying with a urediospore suspension of wheat leaf rust fungi (Puccinia recondita). The wheat seedlings were then transferred to an environment control room to induce the disease. Fourteen days after the inoculation, the wheat seedlings were compared with those in the nontreated plot to calculate the protective value from the area of the disease. The results were evaluated according to the above criteria.

[0213]

For the compounds produced in Examples 29, 43, 53, 56, 57, 59, and 63, the protective value was evaluated as A. These compounds had no phytotoxicity.

[0214]

Test Example 3 Preventive effect against powdery mildew of cucumber

The 20% emulsifiable concentrate prepared in Preparation Example 1 was diluted with water to prepare a test solution having a concentration of 200 ppm. The test solution was applied to stems and leaves of cucumber seedlings (cultivar: Suyo) of first leaf development stage raised in a environment controlled greenhouse. The cucumber seedlings, to which the test solution had been applied, were air dried. Thereafter, the cucumber seedlings were inoculated by spraying with a spore suspension of cucumber powdery mildew fungi (Sphaerotheca fuliginea) to the leaf face. The cucumber seedlings were then transferred to an environment control room to induce the disease. Ten days after the inoculation, the cucumber seedlings were compared with those in the nontreated plot to calculate the protective value from the area of the disease. The results were evaluated according to the above criteria.

[0215]

For the compounds produced in Examples 6, 23, 28, 29, 33, 34, 35, 36, 40, 48, 56, and 71, the protective value was evaluated as A. These compounds had no phytotoxicity.

[0216]

[Advantages of the Invention]

The compound of the present invention is useful for various diseases harmful to agriculture and gardening. In particular, the compound of the present invention has significant control effect against plant diseases, such as rice blast, powdery mildew of cucumber, and wheat leaf rust. Accordingly, the compound of the present invention makes it possible of providing a harmful organism control agent which is usable for plant.

[Document Type] ABSTRACT

[Abstract]

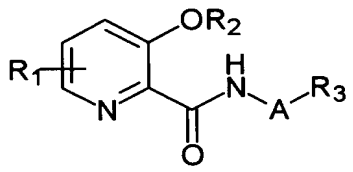
[Object]

To find a picolinamide derivative useful as a harmful organism control agent.

[Means for Solving the Problems]

There is provided a picoliniamide derivative represented by formula (1):

[Chem. 1]



(1)

wherein

[A represents a bond or an optionally substituted alkylene chain; R_1 represents one, two or more groups, which may be the same or different, selected from the group consisting of a hydrogen atom, alkoxy, and haloalkoxy; R_2 represents a hydrogen atom, optionally substituted benzyl, optionally substituted alkyl or optionally substituted alkanoyl; and R_3 represents a hydrogen atom, optionally substituted cycloalkyl, optionally substituted cycloalkenyl, optionally substituted aryl or an optionally substituted heterocyclic group (however, the case where R_1 represents a hydrogen atom, A represents a bond or a methylene chain, and R_3 represents phenyl or cyclohexyl, and the case where A represents an alkylene chain and R_3 represents a hydrogen atom are excluded)].

[Selected Drawing] None